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An Investigation of the Fog Problem in Autoradiographic Intensification Method

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AN INVESTIGATION OF THE FOG PROBLEM IN
AUTORADIOGRAPHIC INTENSIFICATION METHOD

by

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A thesis submitted in partial fulfillment
of the requirements for the degree of
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MASTER'S THESIS

The Master's Thesis of Lawrence E. Wheaton
has been examined and approved
by the thesis committee as satisfactory
for the thesis requirement for the
Master of Science degree

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Submitted to the Photographic Science and Instrumentation Division in partial fulfillment of the requirements for the Master of Science degree at the Rochester Institute of Technology

ABSTRACT

Methods of decreasing the fog density level in an autoradiographically intensified image were investigated. The methods included a more proportional conversion of silver to silver sulfide, decreasing the amount of fog silver in the original image, and minimizing the formation of fog in the original image. Thirty percent exposures of an aluminum 11 step wedge and a lead resolution target were made using Kodak film type XG-1. The underexposed films were either developed in the Kodak RP X-Omat processor or manually developed. All the films were radioactivated in an alkaline solution of sulfur-35 thiourea for 30 minutes. The activated films were exposed to Kodak film type NMC in a radiographic cassette for a specified period of time. The resulting autoradiographs were developed in the X-Omat processor.

A more proportional conversion of silver to radioactive silver sulfide was obtained by increasing the amount of nonradioactive thiourea in the activating solution. The addition of 0.0014 g. of nonradioactive thiourea resulted in a 70% decrease in the fog density of the autoradiograph and a 40% increase in gamma, but at the expense of a 40% decrease in ASA speed. The use of potassium ferricyanide as an oxidizing agent during the radioactivation resulted in increased fog density, while the use of hydrogen peroxide as an oxidizing agent produced little or no effect on the intensification.

Manually developing the underexposed image for a shorter period of time (1 min. instead of 3 min.) resulted in decreased fog from a density of 1.31 to 0.58 and increased contrast from 1.2 to 2.2, while the ASA speed remained unchanged in the intensified image. A fixed-out film which was treated in the photographic subtractive reducer prior to activation did not produce any intensified image; thereby, indicating that sulfur-35 thiourea does not react with the gelatin.

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

I INTRODUCTION

Autoradiographic intensification method (AIM), for the recovery of underexposed images, has recently become a subject for great research activities. AIM involves making an underexposed image radioactive, exposing another emulsion to this radioactivity, and developing the autoradiograph in the conventional manner. The autoradiograph usually gives images with higher densities than the underexposed image. Several methods have been used to make the underexposed images radioactive. These methods include nuclear irradiation of the underexposed images[1,2], radioisotope exchange reaction[1], and radiotoning with one of several radionuclides[1,3]. Recently, Askins introduced a new method of radioactivating the underexposed image by reacting it with an alkaline solution of sulfur-35 thiourea[4,5].

All of the above methods of intensifying an underexposed image have one serious disadvantage in common and that is the non-image silver (fog) is concurrently intensified; thereby, resulting in decreased contrast and loss of image detail in the intensified image. Also important is the fact that the lower density areas (both fog and image) are generally radioactivated more than the higher density areas; thereby, resulting in a disproportionate intensification in the intensified image. It is this disproportionate intensification of the lower density areas that produces increased fog, and decreased contrast and photographic speed, thus decreasing the general quality of the intensified image.

Thackray[1] has indicated several precautions to take to decrease the level of fog in the intensified image by minimizing the formation of the fog in the original image. Vachon[6] treated the underexposed image with a photographic subtractive reducer prior to radioactivation and substantially improved the quality of the autoradiograph by decreasing the fog level and increasing the photographic speed and contrast. However, a reducer does remove some of the image silver and accidental overuse can cause an irreversible loss of image. These methods are applicable to the developed and processed image. Further decreases in the amount of fog in the original image may be obtained by minimizing the formation of emulsion and development fog during development. This may be accomplished by either decreasing development time or using a developer containing a more selective developing agent.

A limited investigation of the cause of the disproportionate radioactivation of the underexposed image has been conducted[7]. The disproportionate radioactivation may be due to a number of factors which include:

1. the use of a high specific activity thiourea solution;
2. the type of oxidizing agent which converts the developed silver (Ag^{o}) to Ag^{+} ion, which then reacts with the thiourea;
3. overdevelopment of the non-image silver; and
4. the reaction between the gelatin and the radioactivating solution.

It is the objective of this thesis to study the extent to which the above factors contribute to the disproportionate radioactivation of the underexposed image and to suggest possible solutions to decreasing the fog and increasing the contrast and photographic speed of the intensified image. For the purpose of radioactivating the underexposed image, sulfur-35 thiourea was used.

II. GENERAL EXPERIMENTAL METHOD

II. A. Donor/Receiver Films

The underexposed original, which is to be intensified, will be referred to as the donor and the film to which it will be exposed, the receiver. The donor and receiver films used throughout the experiments were Kodak film types XG-1, a general radiographic film, and NMC, a nuclear medicine radiographic film, respectively. The XG-1/NMC films were selected as the best donor-receiver based on a survey of radiographic films for autoradiographic intensification by J. Ross[7]. Kodak film type XG-1 is a two-sided, blue based emulsion; NMC is a single-sided, clear based emulsion.

II. B. Production of Donor Films

An 11 step aluminum wedge was calibrated at 80 kilovolts peak (KVp), the KVp setting to be used for the production of all radiographs. The calibration procedure is described in the Appendix. The exposure for which all 11 steps could be easily distinguished was selected as a normal exposure. Several 30% exposures of the step wedge and a lead resolution target were made by keeping the KVp and distance from the tube to the film constant and setting the milliamp-second (mAs) setting at 0.30 times the mAs setting for the normal exposure. The exposures were made on Kodak film type XG-1 in a Kodak X-Omatic regular intensifying screen cassette. Both the normal and underexposed radiographs were processed in a Kodak RP X-Omat processor unless otherwise specified.

II. C. Intensification Method: Activation and Exposure

Sulfur-35 thiourea stock solutions were prepared using five millicurie (mCi) activity thiourea (1.4 mg.) which was dissolved in 250 ml. of distilled water. The activating solution consisting of 25 ml. of the stock thiourea solution, 25 ml. of 0.1 N. sodium hydroxide, and 150 ml. of distilled water was prepared at room temperature immediately prior to use. Any variation in the specific activity was done by adding to it a determined amount of nonradioactive thiourea. At all times the total activity of the solution remained constant at 0.5 mCi. The film to be radioactivated was taped inside a Chromega dual action processing drum and first prewashed with a 50% methanol/water solution for five minutes and then for 20 minutes with four changes of distilled water. Immediately after the washing, the activating solution was poured into the drum and the agitation continued for 30 minutes. Upon completion of the radioactivation, the film was washed in the drum with distilled water for five minutes followed by 20 minutes in a tray of running distilled water. The film was air-dried at room temperature for at least one hour. The radioactivated film was exposed to the receiver film for the desired time, usually two hours, in a radiographic cassette. Processing of the autoradiograph was done in the Kodak RP X-Omat processor.

All of the above intensification procedures were followed unless otherwise specified. All autoradiograph comparisons were made only if activations were done using the thiourea solution of the same lot number.

II. D. Use of Photographic Subtractive Reducer

In some cases, prior to radioactivation, the donor was treated in a photographic subtractive reducer consisting of potassium ferricyanide and sodium thiosulfate. The reducer which was used contained the following:

Solution A	$K_3Fe(CN)_6$	0.2 g.
	distilled water to make	10.0 ml.
Solution B	$Na_2S_2O_3 \cdot 5H_2O$	8.0 g.
	distilled water to make	200.0 ml.

Solutions A and B were mixed just prior to use. Samples to be treated in a reducer were taped at one end to a bottom of a tray. The subtractive reducer was added and the tray was manually agitated for the specified time. The samples were then washed for 20 minutes in running distilled water.

II. E. Measurements

All optical density measurements were obtained using a MacBeth TD 504 transmission densitometer with a 2 mm. aperture. The resolution, in lines per millimeter (lpm.), was determined by viewing the imaged resolution target over a light table with the aid of an eight power magnifier.

The log relative exposure values used for the 30% exposed images were determined by multiplying the exposure values determined in the Appendix (Calibration of the Aluminum 11 Step Wedge) by 0.30. Characteristic curves of the 30% exposed images and the autoradiographs were made by plotting density (D) versus log relative exposure (log H). Gamma, defined as the slope of the straight line portion of the D-log H curve, was used as a measure of contrast. If no straight line portion of the curve existed, the slope of a line tangent to the curve at a point on the middle of the curve was used as the gamma.

The ASA speed was determined using the exposure value corresponding to a density of 0.5 above the base plus fog density. The speed was then expressed as a fractional increase or decrease compared to the 30% exposure. This was determined by taking the ratio of the speed of the intensified image to the speed of the 30% exposed image and was expressed as relative speed increase.

The fog density, which excluded the base density, was also determined for each intensification. This was measured at a place on the film which received no known exposure. The base density of each film was also determined by fixing out an unexposed and unprocessed piece of film. The maximum density recorded was that density on the film which received the exposure from the unattenuated X-ray source. All recorded maximum densities account for the emulsion density only and do not include the density of the film base.

III EFFECT OF INCREASING THE AMOUNT OF NONRADIOACTIVE THIOUREA IN THE ACTIVATING SOLUTION

III. A. Introduction

High specific activity (HSA) thiourea solutions have generally been used to radioactivate underexposed silver images in autoradiographic intensification method (AIM) in order that short contact times may be used[4]. However, only small quantities of thiourea are contained in such HSA thiourea solutions. During the radioactivation step of the autoradiographic intensification process, the sulfur-35 thiourea reacts with both the image and fog silver in the presence of excess hydroxyl ion to produce silver sulfide. Using the area, average density, and covering power of the film samples to be intensified, the amount of silver in one square centimeter of the Kodak film type XG-1 was calculated to be 0.19 mg. Five millicurie (mCi) of the HSA thiourea contained 1.4 mg. of thiourea[8]. A solution containing 0.5 mCi. of the HSA thiourea was used for each activation on film samples containing 218.6 cm^2 in area. That is, 0.14 mg. or 1.8×10^{-6} mole of thiourea was available for the reaction with 43 mg. or 0.4×10^{-3} mole of silver.

According to Haist[9], the reaction of silver ion with thiourea is represented as shown in the following equation:

$$(H_2N)_2C{=}S + 2Ag^+ + 2OH^- \longrightarrow Ag_2S + H_2N{-}C{\equiv}N + 2H_2O$$

That is, for complete conversion of silver to silver sulfide, one mole of thiourea is needed for two moles of silver. As calculated above, the HSA thiourea solution contains an insufficient amount of thiourea to react with all the silver.

To investigate the influence of specific activity on the autoradiographic intensification method, nonradioactive thiourea was added to the HSA thiourea, which was then used for the radioactivation of the underexposed image.

III. B. <u>Experimental Method</u>

The radioactivating solution was prepared by adding 0.14, 1.4 and 14.0 mg. of nonradioactive thiourea to 0.5 mCi. of the HSA thiourea solution to produce solutions of 1.78, 0.32, and 0.03 mCi/mg., respectively. A 3.57 mCi/mg. solution, to which no nonradioactive thiourea was added, was also prepared. The underexposed images used in all these experiments were of an aluminum 11 step wedge and a lead resolution target, and were made as described in section II. B. of this thesis. The film samples were placed in the processing drum, washed, and radioactivated with the appropriate specific activity thiourea solution as described in section II. C. The radioactivated films were exposed to

Kodak film type NMC for two and four hours. Each exposed NMC film was processed in the conventional manner using the Kodak RP X-Omat processor.

Optical densities were measured and characteristic curves were constructed. The resolution in lines per millimeter (lpm.), ASA speed, and gamma of the 30% exposed images and their intensified images were all obtained as described in section II. E.

Based on the results of the intensification experiments using the different specific activity thiourea solutions, radioactivation time series experiments were performed with 0.32 mCi/mg. thiourea. The radioactivation time was varied for 30, 45, and 60 minutes. The intensification process and measurements were performed for each radioactivation time as described previously.

III. C. <u>Results</u>

The characteristic curves for the normal exposure, 30% exposure, and the intensified 30% exposures for the various specific activity solutions are shown in Figures 1 and 2 for exposure time periods of two and four hours, respectively. The photographic parameters of fog density, maximum density, speed increase, gamma, and resolution for these exposures are given in Tables 1 and 2.

As shown in Figures 1 and 2, the decrease in specific activity of the thiourea solution has resulted in a decrease in the fog density of the intensified image, as expected.

For the two hour exposure time, the best specific activity for minimizing the fog density, while maintaining a high maximum density, was 0.32 mCi/mg. (Figure 1, curve 2). For this specific activity, the fog density has been decreased from 0.75 to 0.23 (curves 4 and 2), while the maximum density increased from 2.37 to 2.68. Increasing the exposure time from two to four hours resulted in an intensification with a higher fog density of 0.42 (Figure 2, curve 2), and increased maximum density, speed, gamma, and resolution for the 0.32 mCi/mg. activation. A comparison of these results is shown in Figure 3 and Table 3.

The characteristic curves for the different activation times using the 0.32 mCi/mg. activating solution are shown in Figure 4 and the relevant photographic parameters are given in Table 4. The increased activation time produced an intensified image with a very moderate increment in fog density, maximum density, and ASA speed. Both the gamma and resolution remained almost constant. From a visual inspection of Figure 4, the 45 minute activation time (curve 3) was considered the optimum activation time using 0.32 mCi/mg. sulfur-35 thiourea.

III. D. Discussion

For the complete conversion of the image silver to silver sulfide, two moles of silver were required for every mole of thiourea as previously described by the equation. Since there was 0.4 mg. of silver, 0.2 mg. or 15.2 mg.

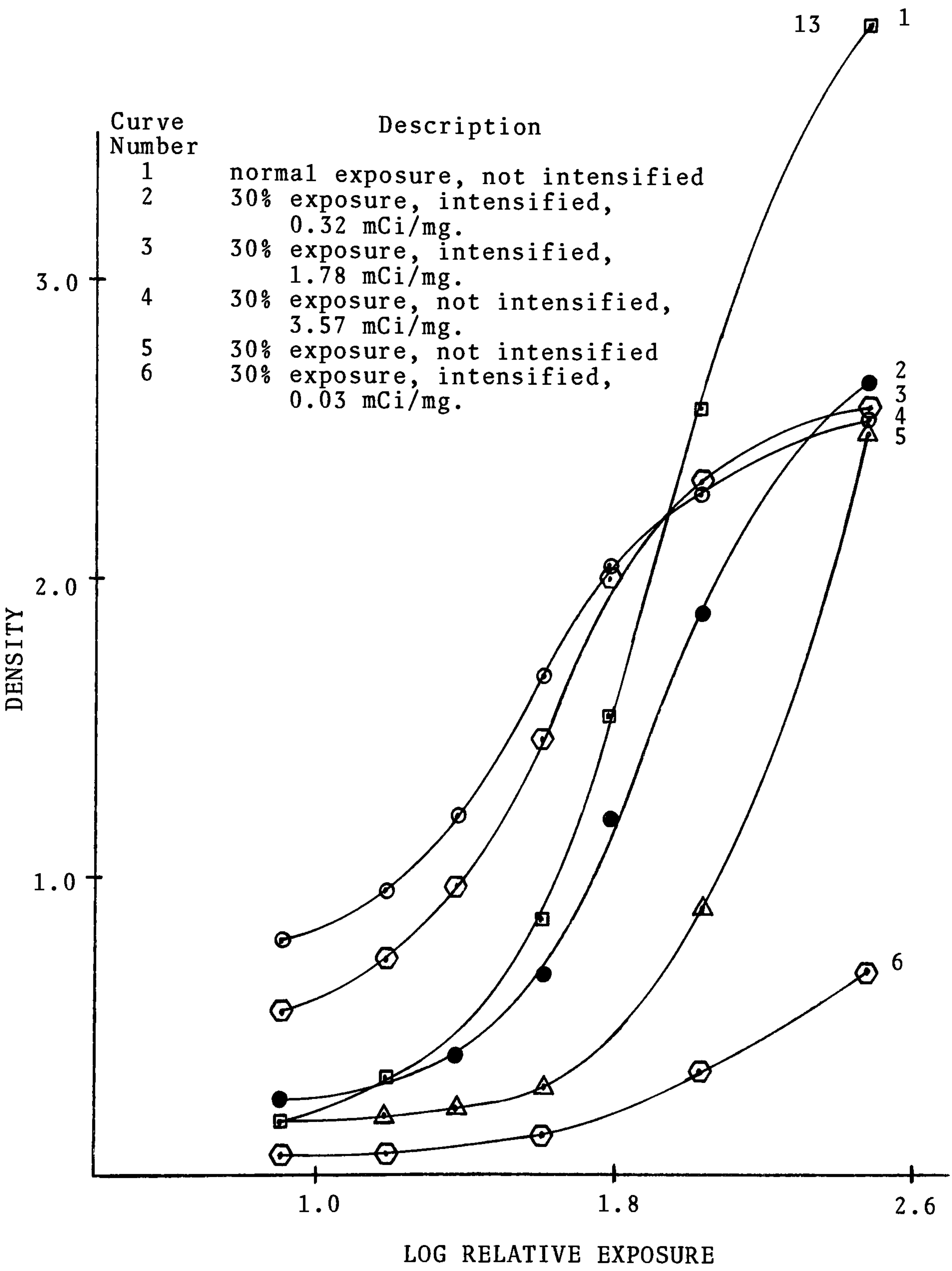


Figure 1. Characteristic Curves of 30% and Normal Exposures and Intensified Images of 30% Exposures Obtained Using a 0.5 mCi. Thiourea Activating Solution of Various Specific Activities and an Exposure Time of Two Hours

TABLE 1

Parameters for 30% Exposed Image and Its Intensified Images Obtained Using Thiourea of Various Specific Activities and Exposed for Two Hours

Photographic Parameter	30% Exposed Image	Intensified Image Specific Activity** (mCi/mg.)			
		3.57	1.78	0.32	0.03
Fog Density*	0.06	0.75	0.57	0.23	0.05
Max. Density*	2.53	2.37	2.58	2.68	0.68
Relative Speed Increase	1.0	3.3	3.1	1.9	0.4
Gamma	2.2	1.5	1.9	2.5	0.5
Resolution (lpm)	5.6	3.5	4.0	4.0	3.5

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included
** Total activity of thiourea solution constant at 0.5 mCi.

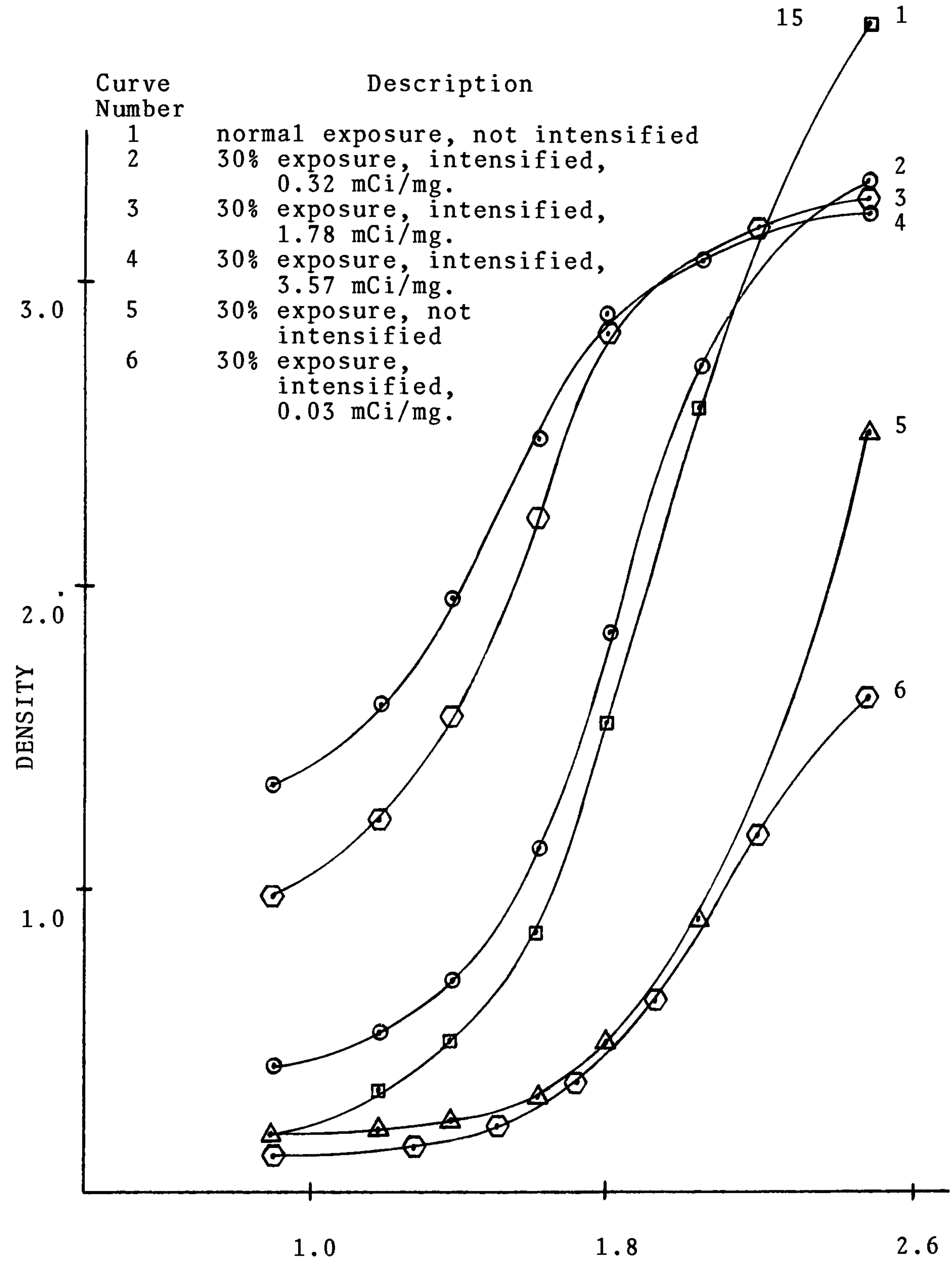


Figure 2. Characteristic Curves of 30% and Normal Exposures and Intensified Images of 30% Exposures Obtained Using a 0.5 mCi. Thiourea Activating Solution of Various Specific Activities and an Exposure Time of Four Hours

TABLE 2

Parameters for 30% Exposed Image and Its Intensified Images Obtained Using Thiourea of Various Specific Activities and Exposed for Four Hours

Photographic Parameter	30% Exposed Image	Intensified Image Specific Activity** (mCi/mg.)			
		3.57	1.78	0.32	0.03
Fog Density*	0.06	1.30	0.98	0.42	0.10
Max. Density*	2.53	3.25	3.29	3.36	1.64
Relative Speed Increase	1.0	4.1	3.9	2.6	1.1
Gamma	2.2	1.7	2.5	2.8	1.4
Resolution (1pm)	5.6	0.0	2.5	4.5	4.0

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included
** Total activity of thiourea solution constant at 0.5 mCi.

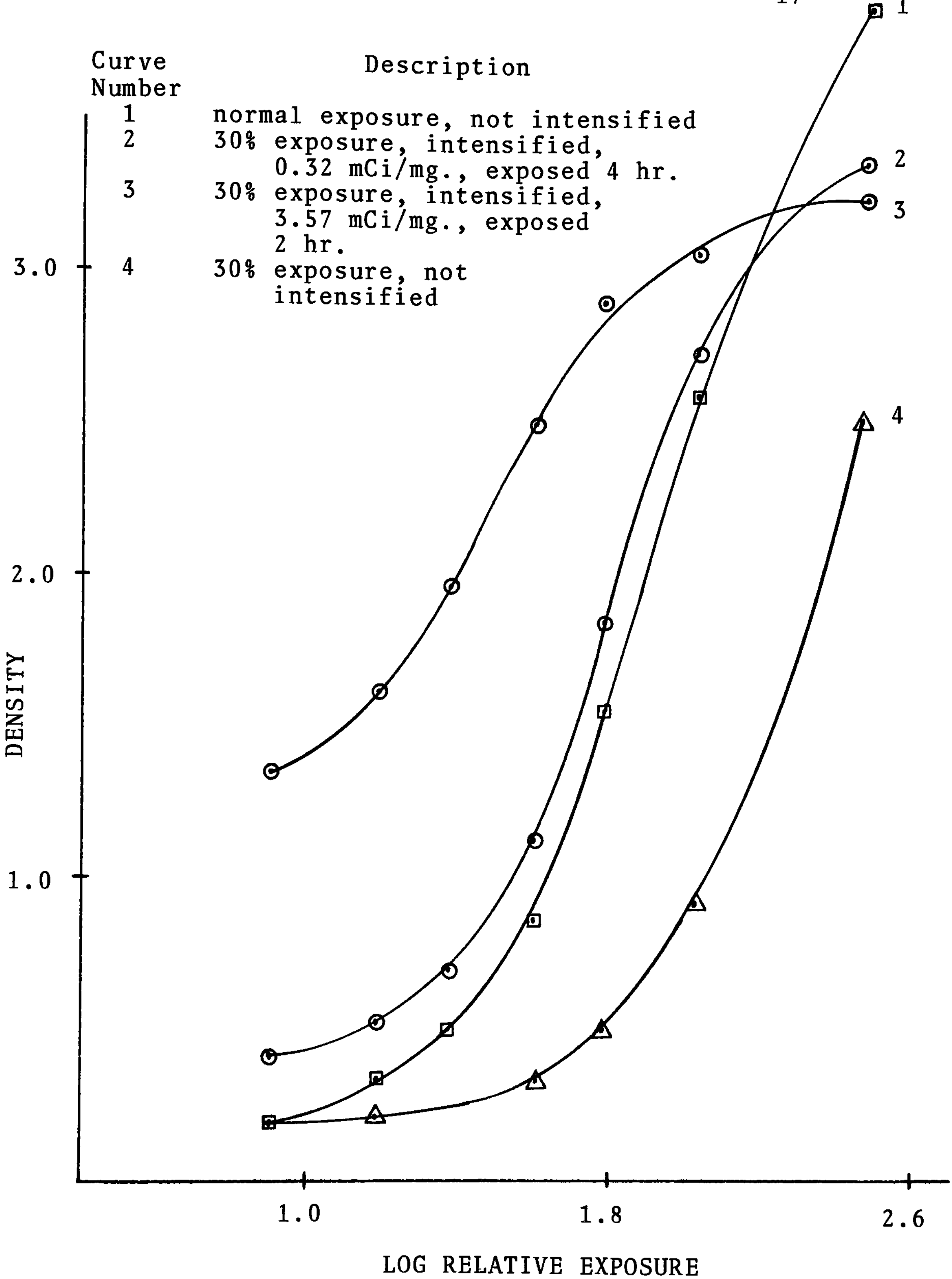


Figure 3. Characteristic Curves of 30% and Normal Exposures and Intensified Images of 30% Exposures Obtained Using a 0.57 and 0.32 mCi/mg. Thiourea and an Exposure Time of Two and Four Hours Respectively

TABLE 3

Comparison of Photographic Parameters of Intensified Images Obtained Using 0.32 mCi/mg. Thiourea and Exposed for Two and Four Hours

Photographic Parameter	Exposure Time 2 hr.	4 hr.
Fog Density* Before Intensification	0.06	0.06
Fog Density* After Intensification	0.23	0.42
Max Density*	2.68	3.36
Relative Speed Increase	1.9	2.6
Gamma	2.5	2.8
Resolution (lpm.)	4.0	4.5

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

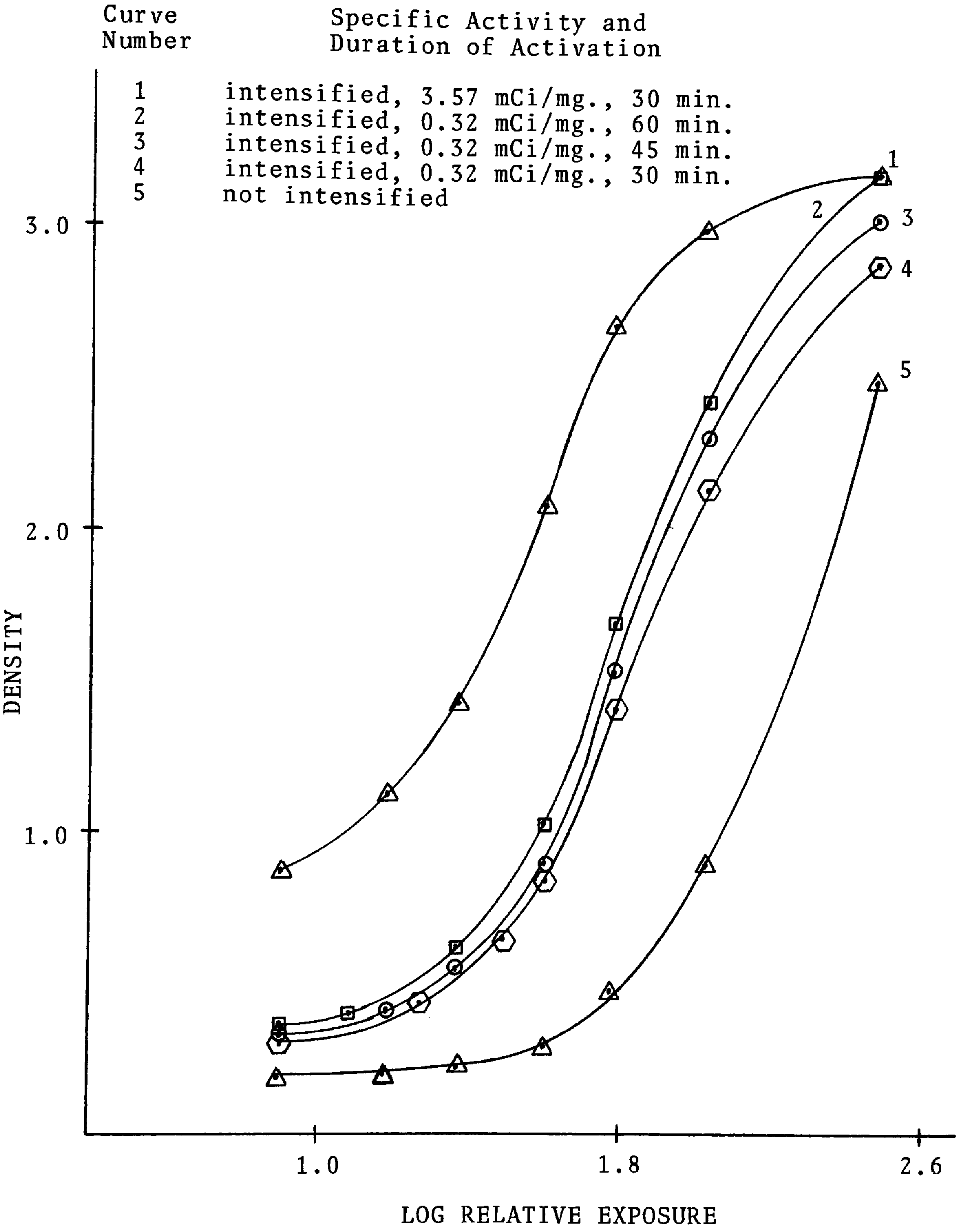


Figure 4. Characteristic Curves of 30% Exposure and Its Intensified Images Obtained by Activating for a Period of 30 min. in 3.57 mCi/mg. Thiourea and for Various Periods of Time in 0.32 mCi/mg. Thiourea and Exposing for Two Hours

TABLE 4

Parameters for 30% Exposed Image and its Intensified Images Obtained by Activating for Various Periods of Time in 0.32 mCi/mg. Thiourea and Exposed for Two Hours

Photographic Parameter	30% Exposed Image	Intensified Image Activation Time (min.) 30	45	60
Fog Density*	0.06	0.29	0.30	0.33
Max Density*	2.35	2.87	3.01	3.15
Relative Speed Increase	1.0	1.9	2.0	2.2
Gamma	2.2	2.1	2.2	2.5
Resolution (lpm.)	5.6	4.0	4.0	3.5

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

of thiourea was required. The 0.03 mCi/mg. thiourea solution contained 14.2 mg. of thiourea and therefore had enough thiourea for an almost total conversion. However, the intensified image from the 0.03 mCi/mg. activation for both the two hour (Figure 1, curve 6) and the four hour (Figure 2, curve 6) exposure had densities, contrast, and ASA speed which were even lower than those for the 30% exposed image. Thus, the 0.03 mCi/mg. intensification produced a degredation in the image quality of the 30% exposed image. This was due to too much nonradioactive thiourea reacting with the silver image.

As the amount of nonradioactive thiourea in the radioactivating solution was decreased from 14.2 mg. (Figure 1, curve 6) to 1.4 mg. (Figure 1, curve 2) and 0.14 mg. (Figure 1, curve 3), the intensified images were characterized by an overall increase in densities. Thus, more of the radioactive thiourea reacted with the silver as the amount of nonradioactive thiourea was decreased. However, if the concentration of nonradioactive thiourea was decreased too much, the fog density increased considerably up to a maximum of 0.75 for the 3.57 mCi/mg. intensification. This increase in the level of intensified fog, as the amount of nonradioactive thiourea was decreased, may possibly be due to the low concentration of thiourea (0.14 mg.) which is sufficient to react with only a small fraction of the silver image. Under these reaction conditions, a large fraction of the silver in the low silver density areas of the film, while only a small fraction of the silver in the high density areas were radioactivated before the reaction was starved for thiourea. Thus, more intensification in the lower density areas resulted, as evidenced by a high fog density.

A compromise between the high intensification and low fog density was obtained using radioactivating solutions of intermediate specific activities of 1.78 mCi/mg. (Figures 1,2; curve 3) and 0.32 mCi/mg. (Figures 1,2; curve 2). Of these intensified images, the one obtained using the 0.32 mCi/mg. radioactivating solution and exposed for four hours (Figure 2, curve 2) produced the best quality image. This characteristic curve (curve 2) is shown in Figure 3 with the normal (curve 1) and 30% (curve 4) exposed images, and the intensified 30% exposed image activated in the standard HSA thiourea (3.57 mCi/mg.) and exposed for two hours (curve 3). The intensified image obtained by radioactivating in the 0.32 mCi/mg. thiourea and exposed for four hours (curve 2) is superior to the standard intensified image obtained using HSA thiourea, since all the photographic parameters improved except for a slight loss in speed from 3.3 to 2.6. When visually compared to the characteristic curve of the normal exposure (Figure 3, curve 1), the 0.32 mCi/mg. characteristic curve closely approximates it, except for a slightly higher fog density and a lower maximum density.

From a visual inspection of the characteristic curves for the different activation times (Figure 4), and examination of the relevant parameters given in Table 4, there was very little gained in the quality of the intensified image by lengthening the radioactivation time. An increase in the activation time from 30 to 60 seconds resulted in a small increase in densities, speed, and gamma; while the resolution remained about the same. However, the 45 minute radioactivation time was considered the optimum time using 0.32 mCi/mg. thiourea. It appears that after 30 minutes reaction time, almost all the thiourea has reacted; and allowing the reaction to continue further, results in only small amounts of additional silver sulfide being formed.

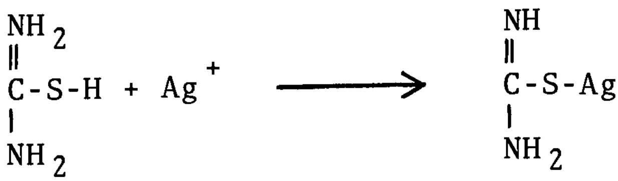
IV. POTASSIUM FERRICYANIDE AND HYDROGEN PEROXIDE AS OXIDIZING AGENTS

IV. A. Introduction

During the radioactivation step of the autoradiographic image intensification process, the thiourea reacts with the image and fog silver to form silver sulfide, as explained below. According to Haist[9], the thiourea (I) is absorbed on the silver halide crystal and converted to the -SH form (II),

(I) (II)

which then reacts with silver ion to form a silver thiourea compound (III).



(II) (III)

In the presence of excess OH^- ion, the silver thiourea (III) reacts with more Ag^+ ion to form the silver sulfide, as shown in the equation below.

$$\overset{\displaystyle NH}{\overset{\|}{C}}\!\!\!\underset{\underset{\displaystyle NH_2}{|}}{}\text{-S-Ag} \xrightarrow[OH^-]{Ag^+} Ag_2S + H_2N\text{-}C{\equiv}N + HOH$$

It is obvious that the image silver (Ag^0) must first be oxidized to silver (Ag^+) ion in order for the above reactions to take place. Since Askins[4] did not introduce an oxidizing agent during the radioactivation process with thiourea, it is assumed that aerial oxygen oxidized the developed silver to silver ion. Due to the limited supply of oxygen, it is further assumed that only a small fraction of the developed silver is oxidized. Increasing the supply of oxygen to the film may overcome this problem but in practice this is a very difficult experiment to carry out. Hence, other oxidizing agents were studied.

The oxidizing agents studied were potassium ferricyanide and hydrogen peroxide. Hydrogen peroxide has a higher oxidizing potential than oxygen, while ferricyanide has a lower potential. Ferricyanide is the most rapid of the well known oxidizing agents for silver[10]. The effects of these oxidizing agents on 30% exposed images, treated and not treated in subtractive reducer, were investigated.

IV. B. Experimental Method

IV. B.1. Potassium Ferricyanide Oxidation

The underexposed images were divided into two sets. One set was treated in the photographic subtractive reducer for one minute prior to radioactivation, while the other set was radioactivated. In either case, the underexposed images were 30% exposures of an 11 step wedge and a lead resolution target, which were made as described in section II.B. To the standard HSA thiourea solution (3.57 mCi/mg.) was added 0.1 g. or 1.0 g. of potassium ferricyanide $K_3Fe(CN)_6$, the oxidizing agent. The film samples were washed, radioactivated in this mixed solution of sulfur-35 thiourea and potassium ferricyanide for 30 minutes in the processing drum, and air-dried, as described in section II. C. The radioactivated film was exposed to Kodak film type NM for two hours and each exposed film was processed in the conventional manner using the Kodak RP X-Omat processor.

Optical densities were measured and characteristic curves were constructed. The resolution, ASA speed, and gamma of the 30% exposed images and their intensified images were all obtained as described in section II. E.

IV. B.2. Hydrogen Peroxide

The same experiments that were done using the potassium ferricyanide as an oxidizing agent were repeated using various concentrations of hydrogen peroxide. The concentrations of hydrogen peroxide studied were 10^{-6} M., 10^{-3} M., and 1.0 M.

IV. C. <u>Results</u>

IV. C.1 Potassium Ferricyanide Oxidation

The characteristic curves for the 30% exposed image and its intensified images, obtained using aerial and potassium ferricyanide oxidation are shown in Figure 5. The corresponding curves for the samples treated for one minute in the subtractive reducer prior to radioactivation are shown in Figure 6. The photographic parameters for these intensifications are recorded in Tables 5 and 6.

The fog density of the intensified, untreated image obtained using aerial oxidation (Figure 5, curve 1) was 0.80, while using 0.1 g. of potassium ferricyanide as the oxidizing agent (Figure 5, curve 3) the fog density was 1.11. The addition of 0.1 g. of potassium ferricyanide also caused the maximum density to decrease from 2.97 to 2.33, the ASA speed to decrease from 3.8 to 3.1, and the gamma to decrease from 2.0 to 1.0. The resolution remained constant at 3.5 lpm. When the amount of potassium ferricyanide was increased to 1.0 g., the intensified step wedge image had a uniform density of 0.73 making it impossible to distinguish one step from the next.

The fog density of the intensified, treated image obtained using aerial oxidation (Figure 6, curve 1) was 0.15, while using 0.1 g. of potassium ferricyanide as the oxidizing agent (Figure 6, curve 2) it was 0.63. The use of 0.1 g. of potassium ferricyanide also produced an increase in the ASA speed from 3.7 to 5.1 and a decrease in the gamma from 2.6 to 1.9.

A comparison of the photographic parameters for the intensified images of the untreated and treated (1 minute in subtractive reducer) underexposures obtained using 0.1 g. of potassium ferricyanide, and the intensified image of the treated underexposure using aerial oxidation is given in Table 7.

IV. C.2. Hydrogen Peroxide Oxidation

The characteristic curves for the 30% exposed image and its intensified image obtained using aerial and hydrogen peroxide oxidation are shown in Figure 7. The corresponding photographic parameters are given in Table 8.

The radioactivation of the donor film using the thiourea solution containing 1.0 M. hydrogen peroxide caused a brown precipitate to form on the donor; and, when exposed to the receiver film, no image was formed. As the concentration of hydrogen peroxide was decreased to 10^{-3} M. and 10^{-6} M., the brown precipitate was not evident on the radioactivated donor film. However, intensified images for both the 10^{-6} M. (Figure 7, curve 1) and the

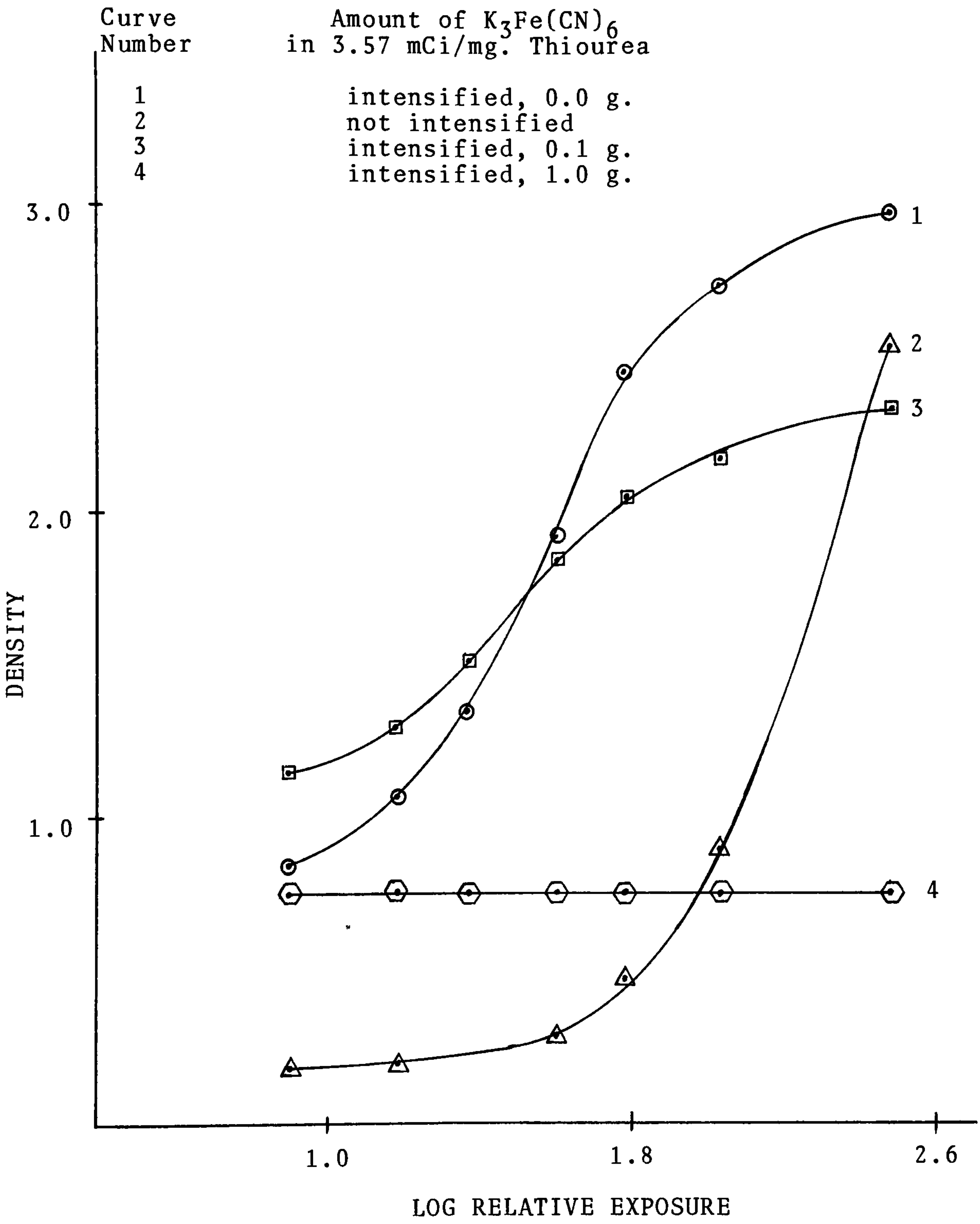


Figure 5. Characteristic Curves of Untreated 30% Exposed Image and Its Intensified Images Obtained Using Various Amounts of $K_3Fe(CN)_6$ in 3.57 mCi/mg. Thiourea and Exposed for Two Hours

TABLE 5

Parameters for Untreated 30% Exposed Image and Its Intensified Images Obtained by Radioactivating in 3.57 mCi/mg. Thiourea Containing Various Amounts of $K_3Fe(CN)_6$ and Exposed for Two Hours

Photographic Parameter	30% Exposed Image	Intensified Image Amount of $K_3Fe(CN)_6$ in Grams		
		0.0 g.	0.1 g.	1.0 g.
Fog Density*	0.06	0.80	1.11	0.73
Max. Density*	2.43	2.97	2.33	0.75
Relative Speed Increase	1.0	3.8	3.1	**
Gamma	2.2	2.0	1.0	0.0
Resolution (1pm)	5.6	3.5	3.5	3.1

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

** Not measureable due to a gamma of zero

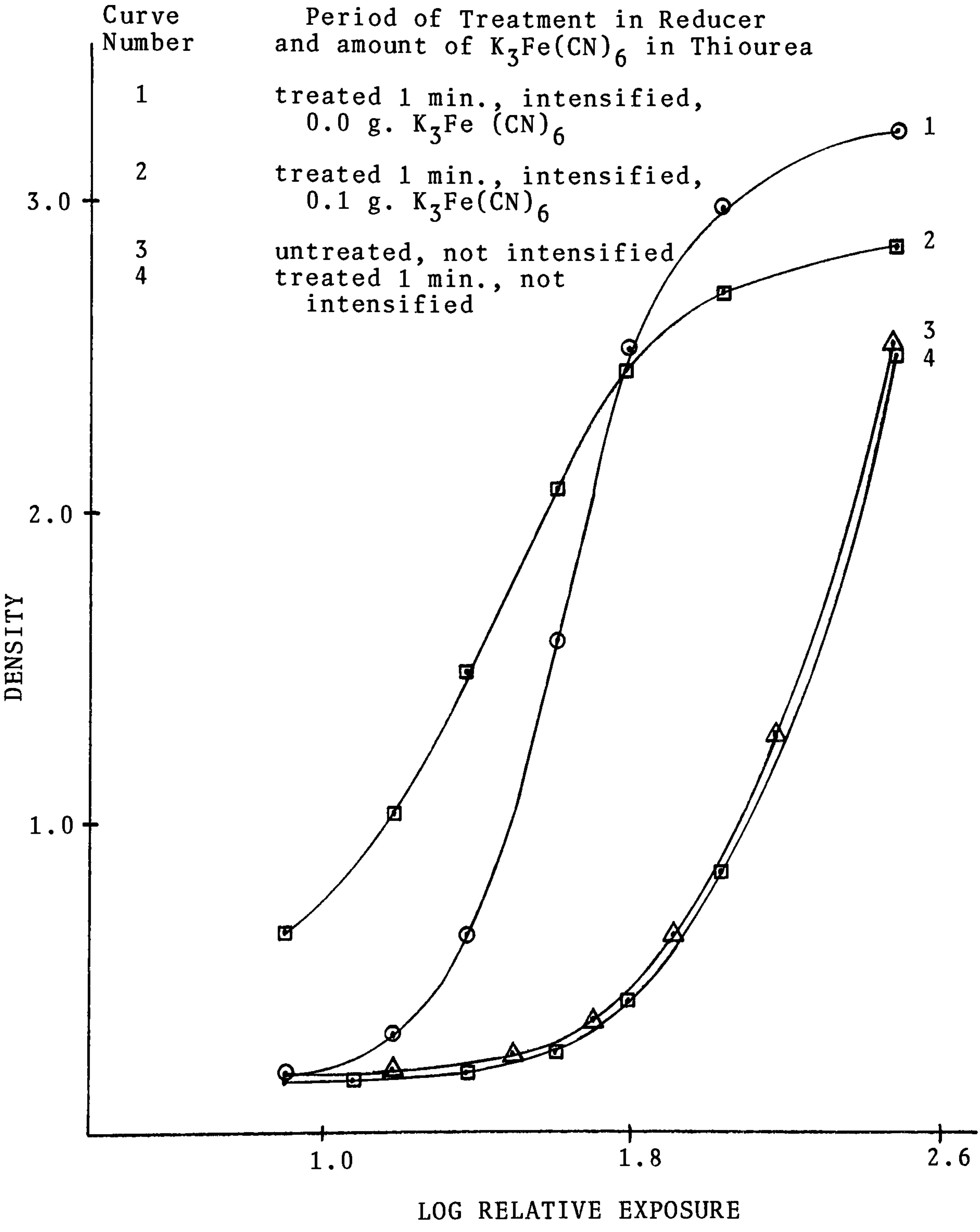


Figure 6. Characteristic Curves of 30% Exposed Image Treated in Subtractive Reducer for 1 Min., and Its Intensified Images Obtained Using Various Amounts of $K_3Fe(CN)_6$ in 3.57 mCi/mg. Thiourea and Exposed for Two Hours

TABLE 6

Parameters for 30% Exposed Image Treated in Subtractive Reducer for 1 Min. Prior to Radioactivating in 3.57 mCi/mg. Thiourea Containing Various Amounts of $K_3Fe(CN)_6$ and Exposed for Two Hours

Photographic Parameter	Treated, 30% Exposed Image	Intensified, Treated Image Amount of $K_3Fe(CN)_6$	
		0.0 g.	0.1 g.
Fog Density*	0.06	0.15	0.63
Max. Density*	2.41	3.23	2.87
Relative Speed Increase	0.9	3.7	5.1
Gamma	2.2	2.6	1.9
Resolution (lpm)	5.6	4.0	4.0

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

TABLE 7

Comparison of Intensified Images of Untreated and Treated (1 Min. in Subtractive Reducer) 30% Exposed Images Obtained by Radioactivating in 3.57 mCi/mg. Thiourea Containing 0.1 g. of $K_3Fe(CN)_6$ and Exposed for Two Hours

Photographic Parameter	Intensified Image		
	$K_3Fe(CN)_6$ Untreated	Oxidation Treated	O_2 Oxidation Treated
Fog Density*	1.11	0.63	0.15
Max. Density*	2.33	2.87	3.23
Relative Speed Increase	3.1	5.1	3.7
Gamma	1.0	1.9	2.6
Resolution (1pm)	3.5	4.0	4.0

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

10^{-3} M. (Figure 7, curve 3) H_2O_2 radioactivations were obtained. Their characteristic curves differ very little from that of the intensified image obtained using aerial oxidation (Figure 7, curve 2).

IV. D. Discussion

IV. D.1. Potassium Ferricyanide Oxidation

The intensified images obtained from using potassium ferricyanide as the oxidizing agent in the radioactivating solution were of poorer quality than those obtained using aerial oxidation. The fog density was higher and the maximum density, speed, and gamma were lower for the image resulting from the addition of 0.1 g. of potassium ferricyanide. This indicates that more of the thiourea was able to react with the low concentration silver in the low density areas of the film and less with the silver in the high density areas. The reason for this occuring may be explained by the rapid oxidation of the silver by the potassium ferricyanide and/or slower diffusion of the fericyanide into the emulsion. It appears that the potassium ferricyanide has oxidized more silver in the low density areas of the film than was oxidized by aerial oxidation. However, the intensified density in the higher density areas was lower for the potassium ferricyanide oxidation than for the aerial oxidation. This lower maximum density may be due to a buildup of silver sulfide on the top layer of the emulsion; thereby, preventing the diffusion of

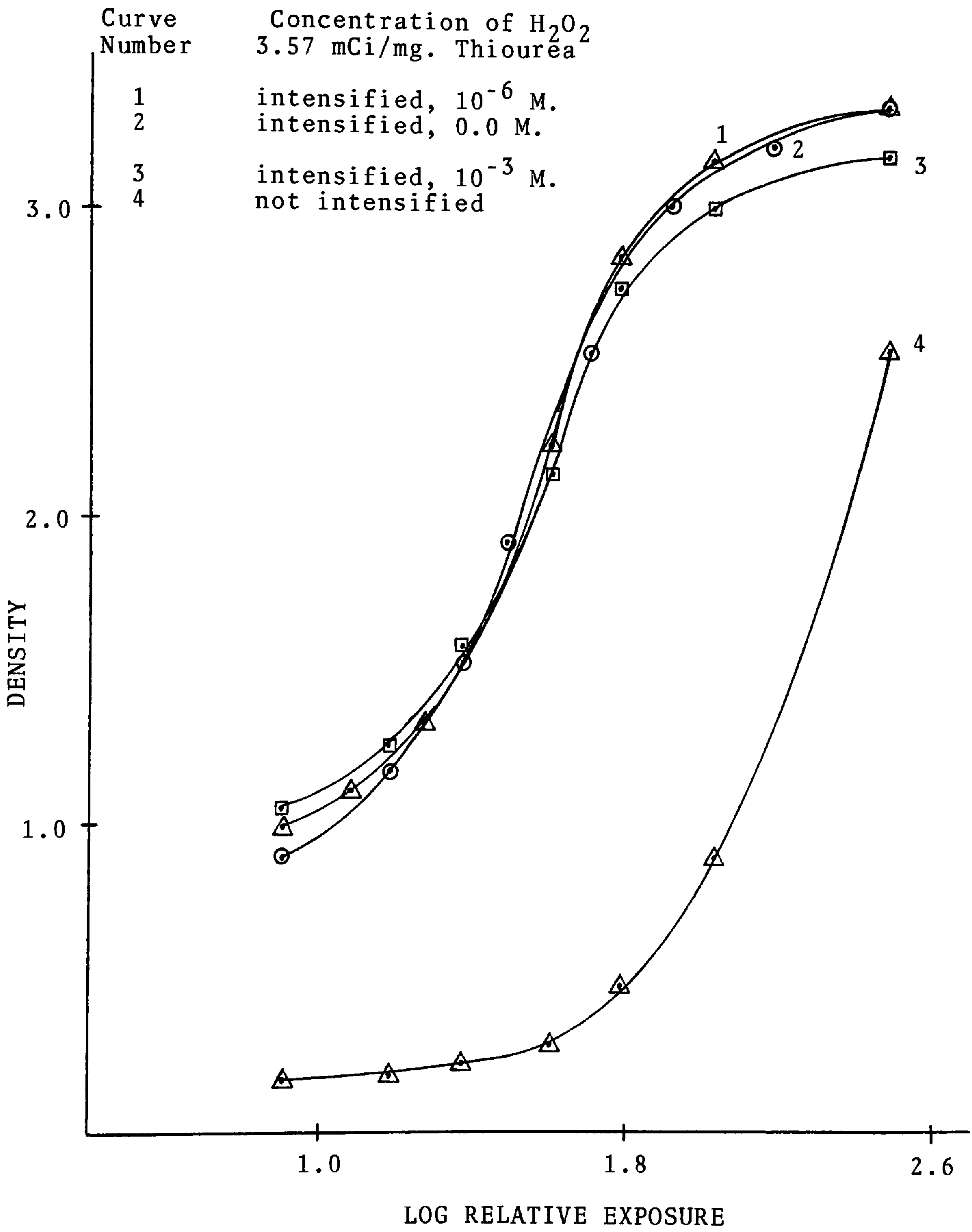


Figure 7. Characteristic Curves of 30% Exposed Image and Its Intensified Images Obtained Using Various Concentrations of H_2O_2 in 3.57 mCi/mg. Thiourea and Exposed for Two Hours

TABLE 8

Parameters for 30% Exposed Image and Its Intensified Images Obtained by Radioactivating in 3.57 mCi/mg. Thiourea Containing Various Amounts of H_2O_2 and Exposed for Two Hours

Photographic Parameter	30% Exposed Image	Intensified Image			
		Concentration H_2O_2 in Thiourea Solution			
		0.0 M.	1.0×10^{-6} M.	1.0×10^{-3} M.	1.0 M.
Fog Density*	0.06	0.87	0.97	1.03	**
Max. Density*	2.43	3.33	3.32	3.16	**
Relative Speed Increase	1.0	4.3	3.9	3.8	**
Gamma	2.2	2.6	2.6	2.5	**
Resolution (lpm)	5.6	3.6	2.8	3.2	**

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

** No intensification obtained

the fericyanide further into the emulsion. This may also explain why a uniform intensified density of 0.73 was obtained when the amount of potassium ferricyanide was increased to 1.0 g.

When the 30% exposed images were treated in the photographic subtractive reducer prior to radioactivation, the fog density decreased from 0.80 to 0.15 for aerial oxidation (Figure 6, curve 1) and from 1.11 to 0.63 for potassium ferricyanide oxidation (Figure 6, curve 2) when 0.1 g. was used. The subtractive reducer decreased the level of intensified fog; but, the intensified image obtained by treating the underexposed image in the reducer and using aerial oxidation is still superior.

The use of potassium ferricyanide has in all cases resulted in an increase in the intensified fog level compared to the level produced by aerial oxidation.

IV. D.2. Hydrogen Peroxide Oxidation

A comparison of the characteristic curves and photographic parameters for the intensified images obtained using hydrogen peroxide and aerial oxygen as the oxidizing agents indicate no significant difference. Since both hydrogen peroxide (10^{-6} M. and 10^{-3} M.) intensifications are almost identical to the aerial oxidation intensification, it can be assumed that the addition of hydrogen peroxide has had no effect. That is, hydrogen peroxide has not oxidized any of the image silver, but all oxidation is still a result of aerial oxidation. On the surface of the image which was radioactivated in the 1.0 M. hydrogen peroxide solution, a brown precipitate of silver oxide was formed, indicating that the peroxide oxidized the silver and formed silver oxide and not silver sulfide. Therefore, no radioactivation took place.

V. TEST FOR THE REACTION OF THIOUREA AND GELATIN

V.A. Introduction

All autoradiographic intensifications, whether from aerial or ferricyanide oxidation, have resulted in the same problem of a greater intensification of the lower densities than the higher ones. In fact, an unexposed and processed emulsion was considerably intensified. Such a fog problem indicates that perhaps the thiourea is reacting with the gelatin to produce a uniform radioactivation and overall fogging in the autoradiograph.

Experiments were conducted to determine if the thiourea does react with the gelatin of Kodak film type XG-1.

V. B. Experimental Method

Unexposed pieces (3 cm. x 4 cm.) of Kodak film type XG-1 were divided into two sets. One set was fixed in a working solution of Kodak rapid fixer for time periods of 1, 5, or 10 minutes to remove the silver halide. This set was further divided into another two subsets. One subset was treated in photographic subtractive reducer (mixture of potassium ferricyanide and sodium thiosulfate) for one minute, as described in section II.D. This subset will be referred to as the fixed-out and treated group. The other subset which was not treated in the reducer will be referred to as the fixed-out group.

The second set of the unexposed films was developed and processed in the Kodak RP X-Omat processor. This set was also subdivided into two subsets. One subset was treated in the photographic subtractive reducer, as described previously, and will be referred to as the processed and treated group. The other subset, which was not treated in the reducer, will be referred to as the processed group.

All four groups - fixed-out, fixed-out and treated, processed, and processed and treated - were radioactivated using an alkaline solution of sulfur-35 thiourea, as described in section II.C. The autoradiographs of each radioactivated film were made in the usual manner. These intensifications were also replicated.

V. C. Results

The fog densities from the two replicates for both the original and intensified images were averaged and are given in Table 9. The fog densities of both the fixed-out, and fixed-out and treated original images were the same (0.01), regardless of the fixation time. However, the densities of their intensified images were significantly different. For the one minute fixation time, the fixed-out density was 0.10, while for the fixed-out and treated it was 0.04. As the period of fixation increased to 10 minutes, the fixed-out density increased to 0.51, while it remained at 0.04 for the fixed-out and treated sample.

The processed image had a density of 0.07 whether or not it was treated in the reducer. However, when the processed images were intensified, a large difference was evident between the densities of the processed (1.88) and the processed and treated (0.33).

V. D. Discussion

The density of 0.07 for the unexposed and processed film suggests that it contained very little silver to react with the thiourea. This low level of optical density, when intensified, resulted in a density of 1.88. Such a high intensification of a low density indicates that perhaps the thiourea had also reacted with the gelatin as well as with the silver during the radioactivation. When the unexposed and processed film was treated in the photographic subtractive reducer prior to radioactivation, the fog density of its intensified image was only 0.33. This result suggests that probably most of the silver in the emulsion had been removed and that intensification was due to either a very small quantity of silver left in the film or due to the reaction of sulfur-35 thiourea with the gelatin. However, when the unexposed film was fixed out and treated in the photographic reducer, the density of the intensified image was only 0.04, compared to a density of 0.01 for the corresponding unintensified image. It is assumed that within the limits of experimental error, that there is no significant difference between the densities of the fixed-out and treated images which were either intensified or not intensified. This result supports the hypothesis that sulfur-35 thiourea does not react with the gelatin and that the fog problem in the intensified image is due primarily to the radioactivation of the non-image silver.

TABLE 9

Average Fog Density* Values for Original and Intensified Samples of Unexposed XG-1 Film, Fixed-Out or X-Omat Processed, and Treated or Not Treated in Subtractive Reducer

Unexposed and Not Processed

	Period of Fixation					
	1 Min.		5 Min.		10 Min.	
	Original A	Intensified B	Original C	Intensified D	Original G	Intensified F
Fixed-out	0.01	0.10	0.01	0.30	0.01	0.51
Fixed-out and Treated	0.01	0.04	0.01	0.04	0.01	0.04

Unexposed and Processed in X-Omat

	Original	Intensified
No Treatment	0.07	1.88
Treated in Reducer 1 Min.	0.07	0.33

* Base densities of 0.12 for XG-1 and 0.02 for NMC are not included

VI. KODAK RAPID FIXER AS A PHOTOGRAPHIC REDUCER

VI. A. Introduction

The treatment of an underexposed image with a photographic subtractive reducer, consisting of potassium ferricyanide and sodium thiosulfate, prior to radioactivation with sulfur-35 thiourea, has been used to decrease autoradiographic fog[6]. The use of fixing baths as photographic reducers has also been investigated[11]. It is therefore possible that Kodak rapid fixer, which is used in the Kodak RP processor, could also be used as a photographic reducer to decrease the fog density in an underexposed image and its autoradiographically intensified image.

The effect of Kodak rapid fixer as a photographic reducer to decrease autoradiographic fog was investigated.

VI. B. Experimental Method

Thirty percent exposed images of an aluminum step wedge and a lead resolution target were made, as described in section II.B., and processed in the X-Omat processor. The underexposed images were treated in Kodak rapid fixer for 5, 10, and 15 minutes or in the subtractive reducer for one minute, followed by a 20 minute wash in running distilled water. One underexposed image was given no treatment in either the reducer or fixer. All images were radioactivated in the processing drum with the HSA thiourea solution for 30 minutes, as described in section II.C., and exposed to Kodak film type NMC for two hours. Each exposed NMC film was processed in the conventional manner using the Kodak RP X-Omat processor.

Optical densities were measured, charcteristic curves constructed, and the relevant photographic parameters determined.

VI. C. Results

The characteristic curves for the 30% exposed image and its intensified images obtained by treating the developed underexposed image in Kodak rapid fixer for 5, 10, and 15 minutes, prior to radioactivation in HSA thiourea, are shown in Figure 8. The photographic parameters for the unintensified and intensified images are given in Table 10.

As the treatment period in the Kodak rapid fixer increased, both the fog and maximum density decreased in each of the original and intensified images. The fog density of the original (0.06) decreased to 0.03 when treated for 15 minutes in the rapid fixer. The corresponding decrease in the fog densities of the intensified image was from 0.84 to 0.07. As the period of fixation increased, the ASA speed of the intensified image decreased from 3.2 for the 5

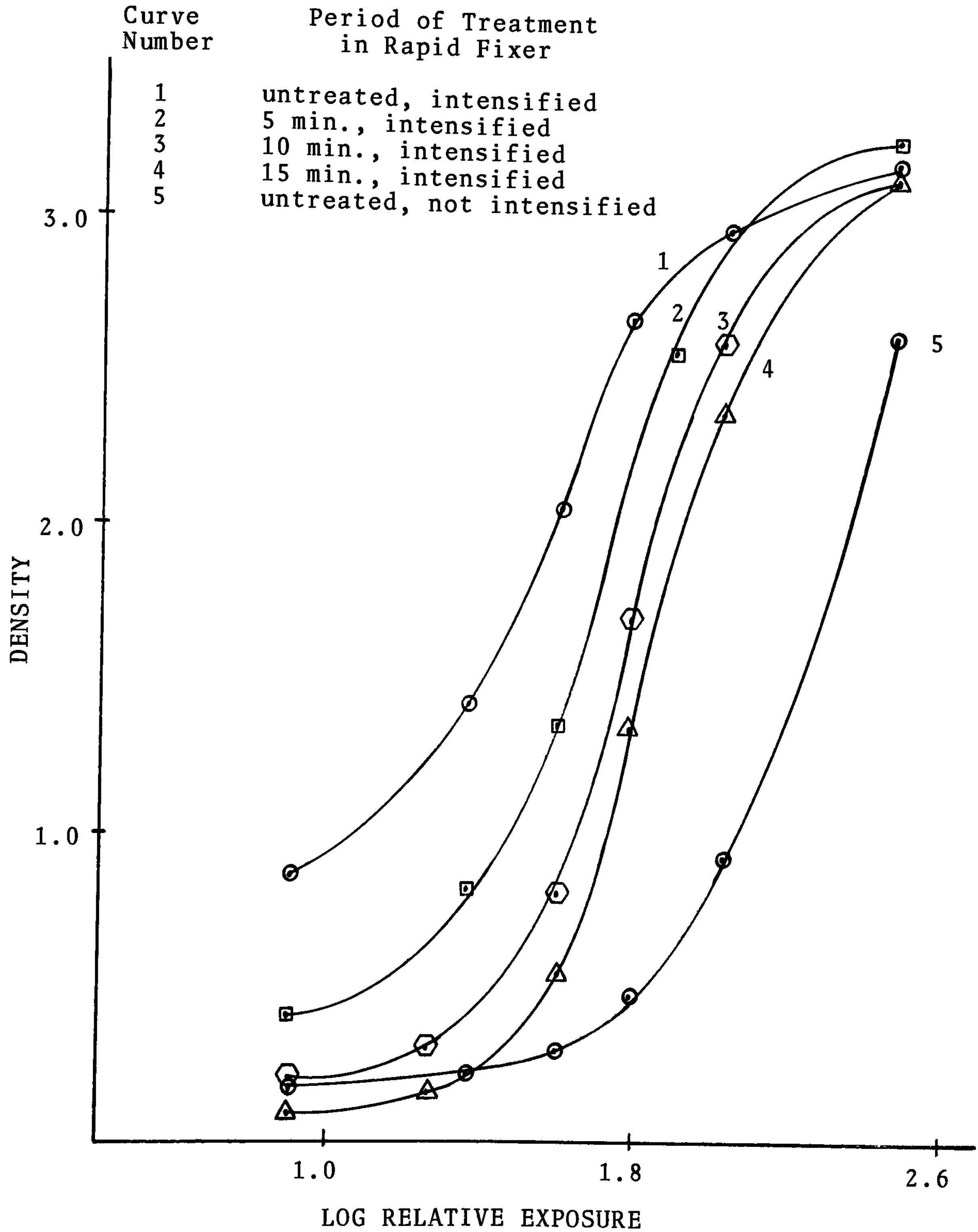


Figure 8. Characteristic Curves of 30% Exposed Image and Its Intensified Images Resulting From Treatment in Kodak Rapid Fixer for 5, 10, and 15 Min., Radioactivation in 3.57 mCi/mg. Thiourea, and Exposed for Two Hours

TABLE 10

Parameters for 30% Exposed Images Pretreated in Kodak Rapid Fixer for Various Periods of Time and Their Intensified Images Obtained by Activating in 3.57 mCi/mg. Thiourea and Exposed for Two Hours

30% Exposed Image

Photographic Parameter	Untreated	Treated			
		Reducer	Rapid Fixer		
		1 min.	5 min.	10 min.	15 min.
Fog Density*	0.06	0.03	0.06	0.05	0.03
Max. Density*	2.51	2.45	2.44	2.33	2.23
Relative Speed Increase	1.0	1.0	1.0	1.0	1.0
Gamma	2.2	2.2	2.2	2.2	2.2
Resolution (lpm)	5.6	5.6	5.6	5.6	5.6

Intensified Image

Photographic Parameter	Untreated	Treated			
		Reducer	Rapid Fixer		
		1 min.	5 min.	10 min.	15 min.
Fog Density*	0.84	0.09	0.38	0.16	0.07
Max. Density*	3.17	3.12	3.23	3.13	3.13
Relative Speed Increase	4.1	3.1	3.2	2.5	2.0
Gamma	2.5	4.0	3.0	3.8	3.8
Resolution (lpm)	3.2	3.6	3.2	4.0	4.0

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

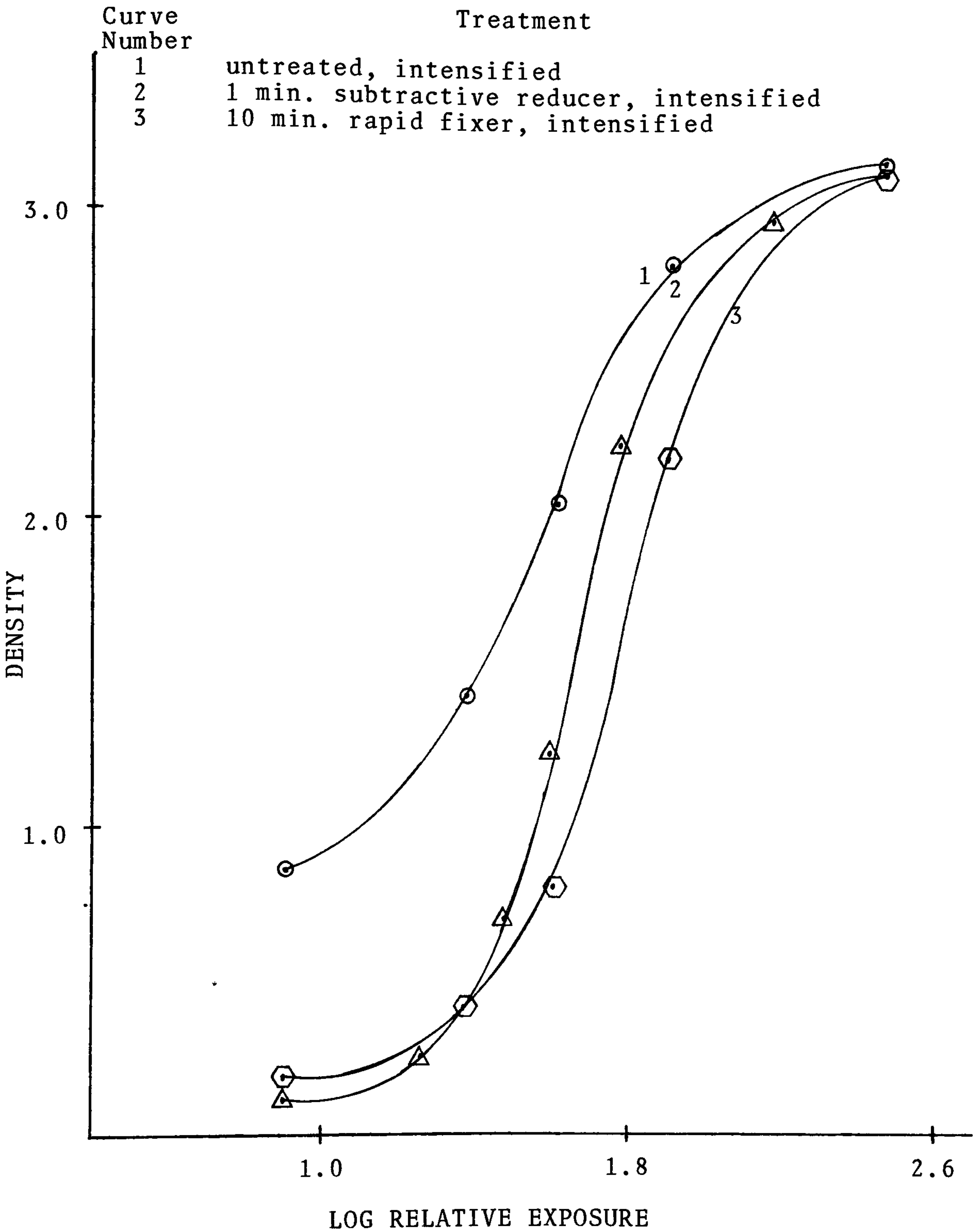


Figure 9. Characteristic Curves of 30% Exposed Images With No Treatment, Treated for 1 Min. in Subtractive Reducer, and Treated for 10 Min. in Kodak Rapid Fixer and Exposed for Two Hours

minute treatment to 2.0 for the 15 minute treatment, while the gamma increased from 3.0 to 3.8. There was no significant change in either the maximum density or the resolution.

The D-Log H curve for the intensified 30% exposed image pretreated in the subtractive reducer for one minute (curve 2) is shown in Kodak rapid fixer for 10 minutes (curve 3).

VI. D. Discussion

The values of the photographic parameters for the 30% exposed images treated in the Kodak rapid fixer indicate that the fixer has performed as a photographic reducer to decrease both the fog and maximum densities. The one minute treatment in the subtractive reducer decreased the fog density from 0.06 to 0.03 and the maximum density from 2.51 to 2.45; while the 15 minute treatment in Kodak rapid fixer produced the same decrease in the fog density but a larger decrease in the maximum density from 2.51 to 2.23. This indicates that perhaps the rapid fixer is not as effect as the subtractive reducer (potassium ferricyanide and sodium thiosulfate) in selectively decreasing the lower densities more than the higher densities. However, when these images were intensified the maximum densities for the one minute reducer and 15 minute fixer treated images were almost the same.

The intensified image of the 30% exposed image treated for 10 minutes in rapid fixer was chosen as the optimum treatment time due to its combined high ASA speed (2.5) and low fog density (0.16). This characteristic curve (curve 3) is shown with the characteristic curve of the intensified, one minute reducer-treated image (curve 2) in Figure 9. Both curves are very similar, but the reducer-treated curve is the better of the two, due to its slightly lower fog density and higher speed.

VII. EFFECT OF VARYING THE DEVELOPMENT TIME OF THE UNDEREXPOSED IMAGE

VII. A. Introduction

So far all experiments have been directed toward treating the donor film after it is developed to minimize the amount of fog. A different approach is to prevent the formation of the fog silver in the first place. One of the major contributors of fog, other than stray radiation, is due to the type of developor used and the length of development.

According to T.H. James[12] there are two types of fog which may be formed from the reaction of the developing agent with the silver halide emulsion:

1. Developer fog which is produced as a result of the developing agent attacking silver ions in solution or the unnucleated silver halide surface, and
2. Emulsion fog which is formed as a result of the catalytic action of nuclei, such as silver sulfide, formed during the ripening stage of the emulsion making process.

Studies by R.W. Swenson[13] of developer and emulsion fog have shown that as development time increases the fog density increases, with the emulsion fog increasing at a faster rate than the developer fog. In some cases, this developer fog may not be detected as transmission density since it has a low covering power, but when reacted with sulfur-35 thiourea it may be radioactivated and does visibly appear as fog in the autoradiograph. Based on this, it would seem reasonable that, since the donor is only a 30% exposure, developing it for the full time recommended for a normal exposure would be overdevelopment. Thus, unnecessary fog would be produced in the donor which would react with the sulfur-35 thiourea to produce excessive fog in the intensified image.

Hence in this section, the underexposed image will be developed for various periods of time shorter than the recommended time, and intensified in the usual manner.

VII. B. Experimental Method

Thirty percent exposed images of the aluminum step wedge and lead resolution target were produced using Kodak film type XG-1, as described in section II.B. The exposed, but unprocessed, films were divided into two sets. One set was developed as usual in the Kodak RP X-Omat processor. Using different development times, the other set was manually processed in photographic trays according to the following procedure:

1. Development was carried out using a working solution of Kodak RP X-Omat Developer Replenisher at 26.5°C. for the designated time with no agitation.
2. Rinsing of the developed film, before fixation, was done in Kodak Stop Bath SB-1a at 26.5°C. for 20 seconds using continuous tray rock agitation.
3. Fixation was accomplished with Kodak RP X-Omat Fixer and Replenisher at 26.5°C for two minutes - one minute in each of two baths - using continuous tray rock agitation.
4. Washing of the film was done in running water at approximately 20°C.
5. Drying of the film was done in a dust-free cabinet at 48°C.

The development times studied were ½, 1, 1½, and 3 minutes. These development donors were radioactivated using the standard HSA sulfur-35 Thiourea activating solution with a specific activity of 3.57 mCi/mg. and exposed on Kodak film type NMC for one and two hours, as described in section II.C.

Based on the results of the above experiments, the one minute development time was considered adequate for developing the 30% exposed aluminum step wedge. The effect of using low specific activity (LSA) thiourea (0.32

mCi/mg.), subtractive reducer, and Kodak rapid fixer on the manually processed (one minute development) and X-Omat processed, underexposed images was investigated. When the subtractive reducer was used, the underexposed images were treated in the solution for various periods of time after complete processing but before radioactivation. When the rapid fixer was used as a reducer, the underexposed image was developed and then fixed in Kodak rapid fixer for the various times. In other words, the rapid fixer was used during the processing of the underexposed image and the subtractive reducer after complete processing.

VII. C. Results

The characteristic curves for the X-Omat and manually processed 30% exposed images and their intensified images using the HSA thiourea (3.57 mCi/mg.) solution are shown in Figure 10. The corresponding photographic parameters are listed in Table 11. As shown in the curves for the unintensified 30% exposed images (curves 5, 6, 7, and 8), as the developmen time decreases the optical density decreases. This decrease in density was very small for the fog density and large for the maximum density. However, when the images were intensified (curves 1, 2, 3, and 4), the maximum densities remained about the same, while the fog densities decreased considerably. For example, the fog density of the intensified image which was processed in the X-Omat (curve 1) was 1.31 and for the image manually processed for a

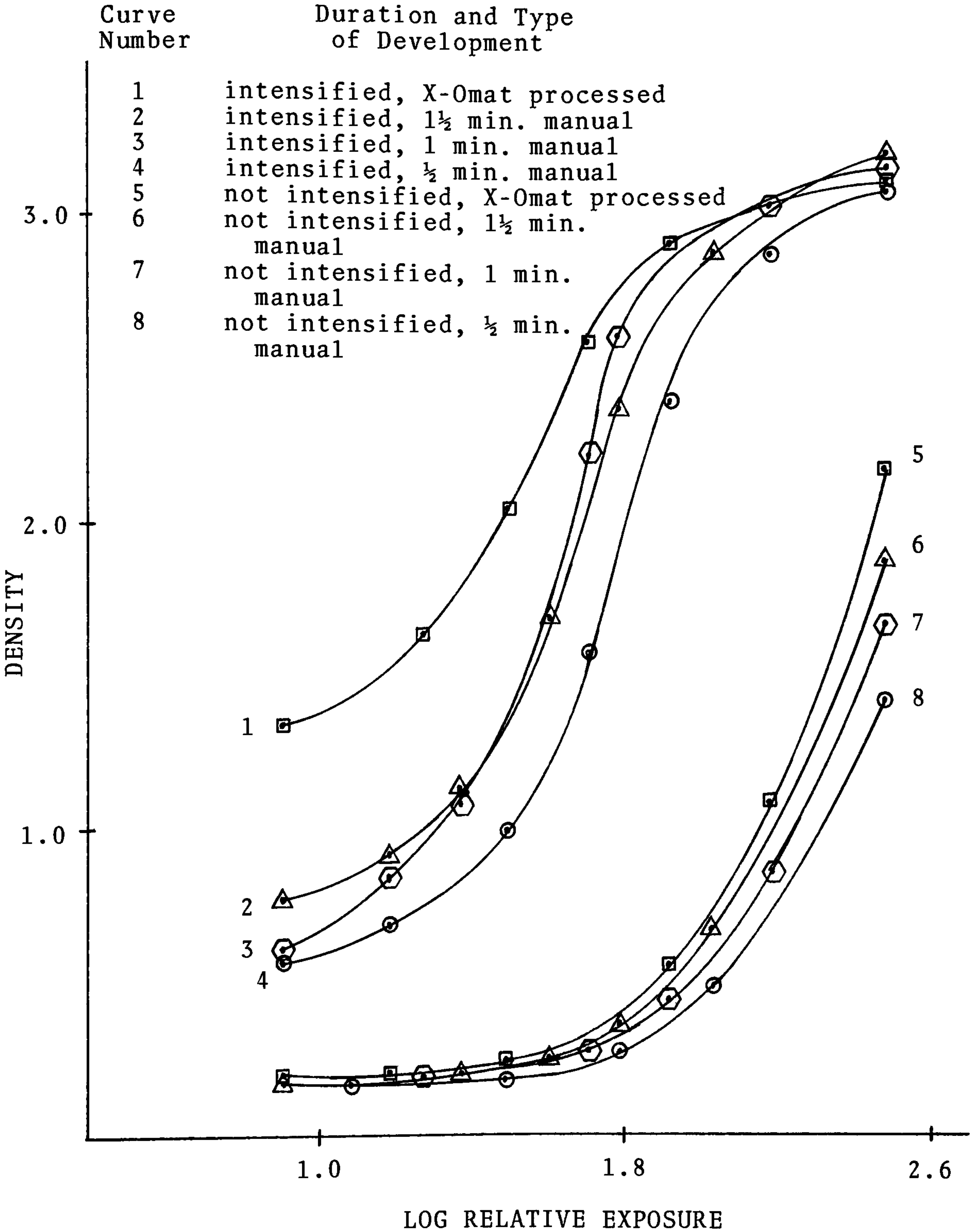


Figure 10. Characteristic Curves of 30% Exposed Images Processed in X-Omat and Manually Processed for Various Development Times and Their Intensified Images Obtained Using 3.57 mCi/mg. Thiourea and Exposed for Two Hours

TABLE 11

Parameters for 30% Exposed Images Developed in the X-Omat or Manually Processed for Various Periods of Development, and Their Intensified Images Obtained Using 3.57 mCi/mg. Thiourea and Exposed for Two Hours

30% Exposed Image

Photographic Parameter	Type and Period of Development			
	X-Omat	Manual		
		½ Min.	1 Min.	1½ Min.
Fog Density*	0.07	0.05	0.05	0.05
Max. Density*	2.07	1.30	1.56	1.75
Relative Speed Increase	1.0	0.6	0.8	0.9
Gamma	2.2	1.7	1.9	2.0
Resolution (lpm)	5.6	5.0	5.6	5.6

Intensified Image

Photographic Parameter	Type and Period of Development			
	X-Omat	Manual		
		½ Min.	1 Min.	1½ Min.
Fog Density*	1.31	0.55	0.58	0.76
Max. Density*	3.10	3.08	3.15	3.21
Relative Speed Increase	3.8	2.8	4.0	3.3
Gamma	1.7	2.7	3.0	2.6
Resolution (lpm)	3.6	4.5	4.0	4.5

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

development time of one-half minute (curve 4) it was 0.55. The intensified image of the one minute developed image was considered to have the best results with a fog density of 0.58, contrast of 3.0, and the highest speed increase of 4.0. In this particular example, the fog density has been decreased from 1.31 to 0.58.

The characteristic curves for the 30% exposed image, developed for one minute, and its intensified image using LSA thiourea (0.32 mCi/mg.) are shown in Figure 11 and the corresponding photographic parameters in Table 12. The use of LSA thiourea (curve 3) produced a small increase in the fog density from 0.05 to 0.12 and an increase in all other parameters, except resolution which decreased from 5.6 to 4.0.

The effects of pretreating the one minute manually developed, underexposed image in the subtractive reducer or using extended fixation in Kodak rapid fixer during manual processing are shown in Figures 12 and 13, respectively; and the corresponding parameters are shown in Tables 13 and 14, respectively. Treating the image in the subtractive reducer for one minute, prior to radioactivation in 3.57 mCi/mg. thiourea, resulted in a decrease in the intensified fog from 0.83 (Figure 12, curve 1) to 0.26 (Figure 12, curve 4), and the ASA speed from 3.3 to 3.2, while the gamma increased from 2.3 to 3.3. Extending the period of fixation in Kodak rapid fixer from 2 to 10 minutes produced a decrease in the intensified fog density from 0.80 (Figure 13, curve 1) to 0.40 (Figure 13, curve 4), while the ASA speed and gamma increased slightly from 2.9 to 3.3 and 3.2 to 3.3, respectively. In both cases the resolution remained unchanged.

VII. D. Discussion

The manual processing procedure outlined in section VII.B. was recommended by Kodak for emergency manual processing of Kodak film type XG-1[14]. To obtain an image similar to that provided by the Kodak RP X-Omat processor, a development time of three minutes was suggested. That is, the 1 1/2 minute development time was 50% of the recommended full development time.

Decreased development resulted in a decrease in the intensified fog densities from 1.31 to 0.55, 0.58, and 0.76 for development times of 1/2, 1, and 1 1/2 minutes, respectively. This decreased fog density may be due to a decrease in the amount of image silver and not a decrease in the amount of developer fog. However, the decreased development did not produce a decrease in the optical fog density of the original. This indicates that possibly less developer fog, which is slow to form and has low covering power, may have been produced[15]. In addition, the development of less emulsion fog may have contributed to the decreased fog levels.

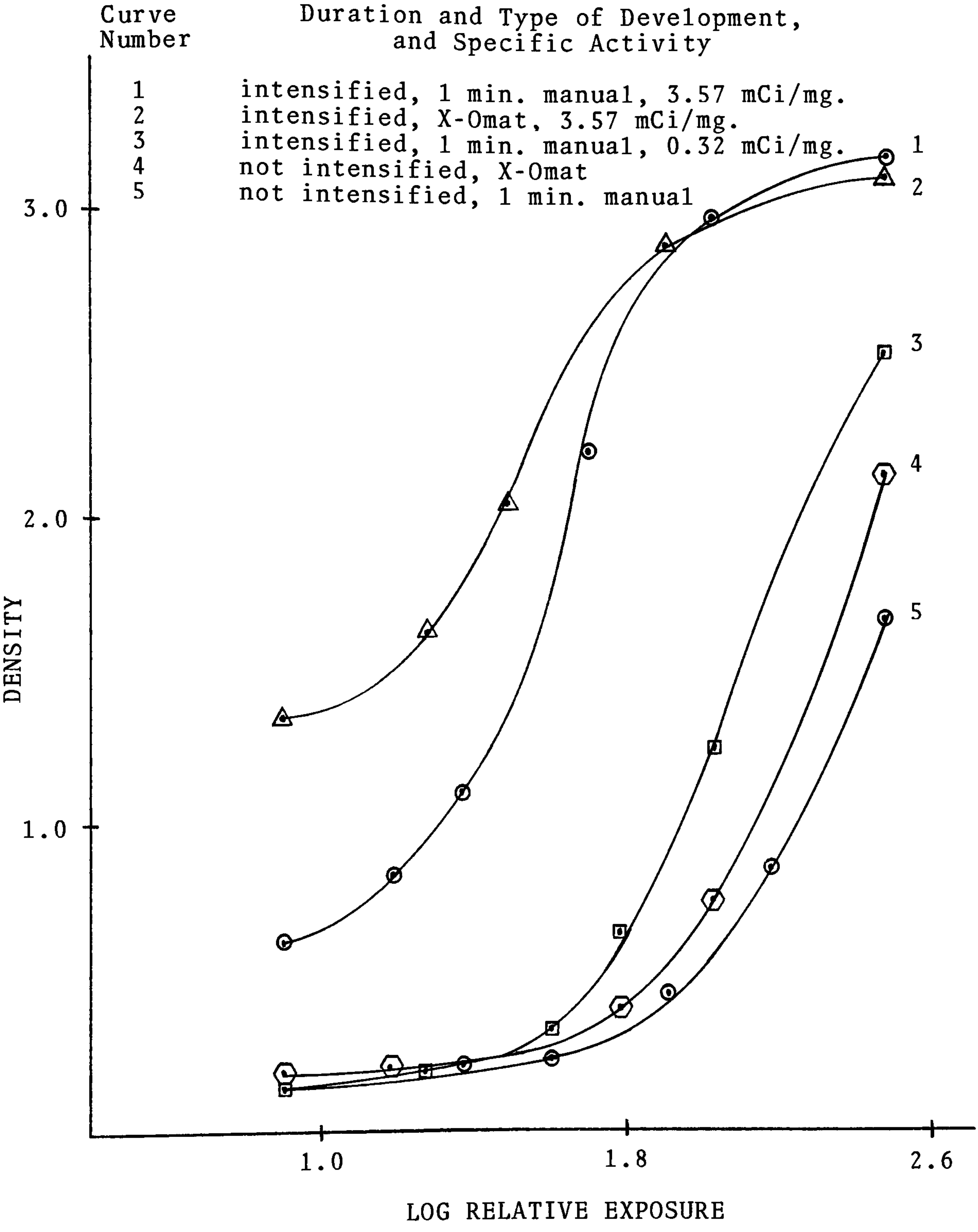


Figure 11. Characteristic Curves of 30% Exposed Images Processed in X-Omat and Manually Processed for Development Time of 1 Min., and Their Intensified Images Obtained Using 3.57 or 0.32 mCi/mg. Thiourea and Exposed for Two Hours

TABLE 12

Parameters for 30% Exposed Images Processed in the X-Omat or Manually Processed for Development Time of One Minute, and Their Intensified Images Obtained Using 3.57 or 0.32 mCi/mg. Thiourea and Exposed for Two Hours

Photographic Parameter	Type and Period of Development, Specific Activity				
	X-Omat		Manual, 1 Min.		
	Original	Intensified 3.57 mCi/mg.	Original	Intensified 3.57 mCi/mg.	Intensified 0.32 mCi/mg.
Fog Density*	0.07	1.31	0.05	0.58	0.12
Max. Density*	2.07	3.10	1.56	3.15	2.51
Relative Speed Increase	1.0	3.8	0.8	4.0	1.4
Gamma	2.2	1.7	1.9	3.0	2.4
Resolution (1pm)	5.6	3.6	5.6	4.0	4.0

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

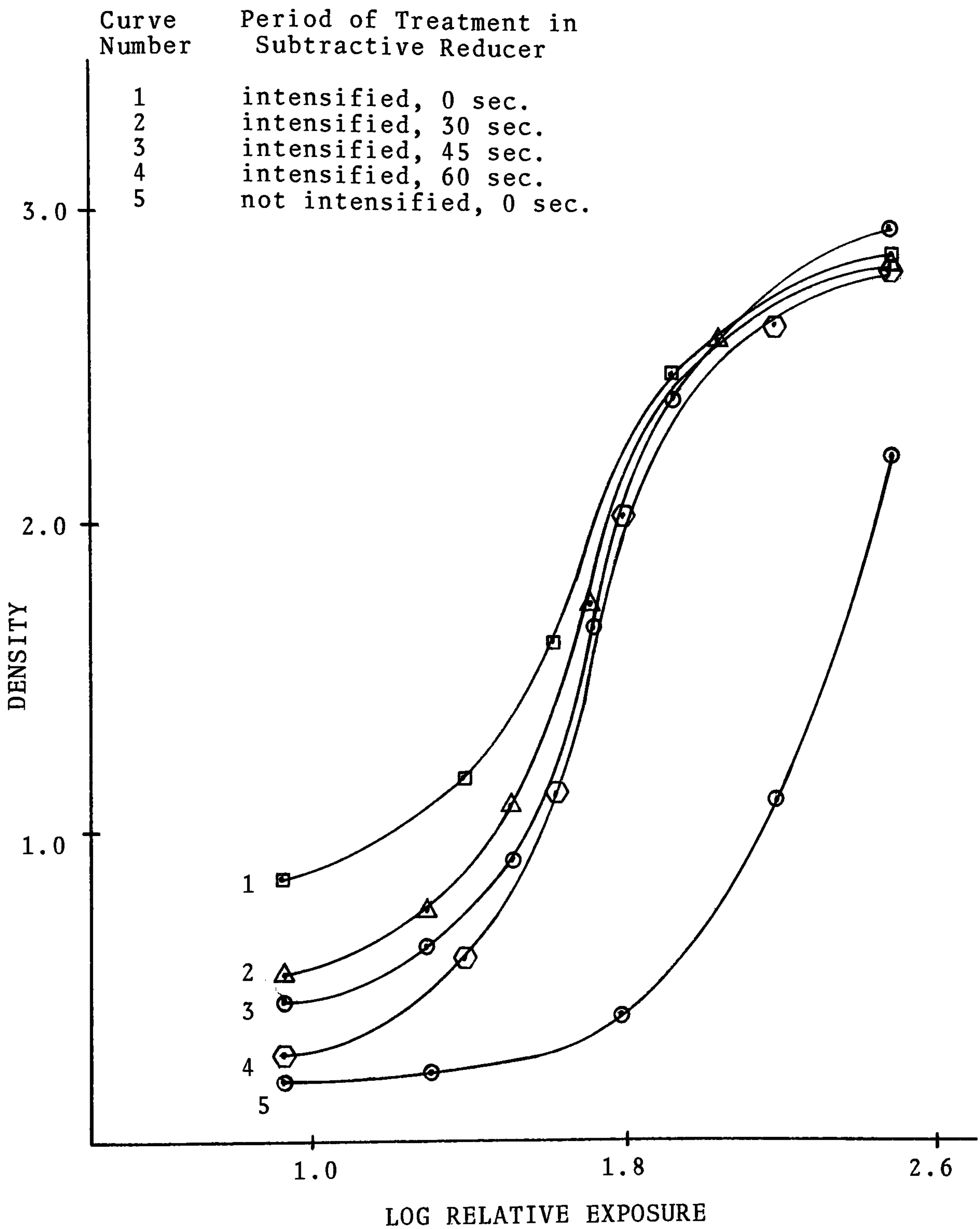


Figure 12. Characteristic Curves of 30% Exposed Image, Manually Processed for Development Time of 1 Min., and Its Intensified Images Obtained by Treating in Subtractive Reducer for 1 Min. Prior to Radioactivation in 3.57 mCi/mg. Thiourea and Exposed for Two Hours

TABLE 13

Parameters for 30% Exposed Image Manually Developed for One Minute, and Its Intensified Images Obtained by Treating in Subtractive Reducer for Various Periods of Time, Activating in 3.57 mCi/mg. Thiourea, and Exposing for Two Hours

Photographic Parameter	30% Exposed Image	Intensified Image			
		Period of Treatment in Reducer			
		0 sec.	30 sec.	45 sec.	60 sec.
Fog Density*	0.08	0.83	0.54	0.43	0.26
Max. Density*	2.10	2.85	2.84	2.93	2.79
Relative Speed Increase	1.0	3.3	3.2	3.1	3.2
Gamma	2.2	2.3	3.0	3.2	3.3
Resolution (lpm)	5.0	4.0	4.0	4.5	4.5

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

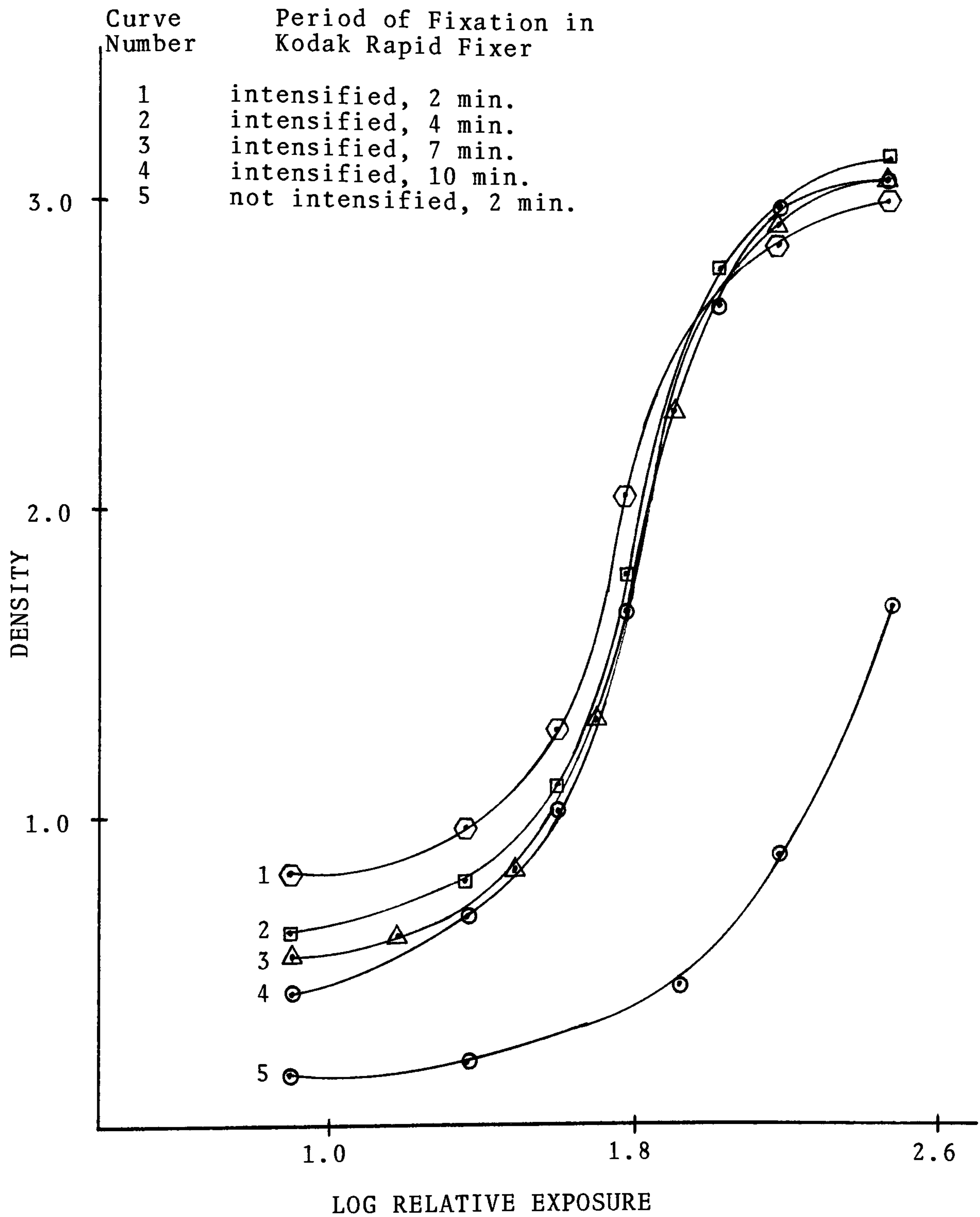


Figure 13. Characteristic Curves of 30% Exposed Image, Manually Processed for Development Time of One Minute and Fixed for Two Min.; and the Intensified Images of the 1 Min. Developed Images Obtained by Treating in Rapid Fixer for Various Periods of Time and Exposed for Two Hours

TABLE 14

Parameters for 30% Exposed Image, Manually Developed for One Minute and Fixed for Two Minutes, and the Intensified Images of 30% Exposed Images Fixed in Rapid Fixer for Various Periods of Time, Radioactivated in 3.57 mCi/mg. Thiourea, Exposing for Two Hours

Photographic Parameter	30% Exposed Image Fixed 2 Min.	Intensified Image			
		Period of Fixation			
		2 Min.	4 Min.	7 Min.	10 Min.
Fog Density*	0.05	0.80	0.59	0.52	0.40
Max. Density*	1.56	2.89	3.14	3.07	3.06
Relative Speed Increase	1.0	2.9	3.0	3.0	3.3
Gamma	1.7	3.2	3.3	3.5	3.3
Resolution (lpm)	5.6	4.5	4.5	4.5	4.5

* Base density of 0.12 for XG-1 and 0.02 for NMC are not included

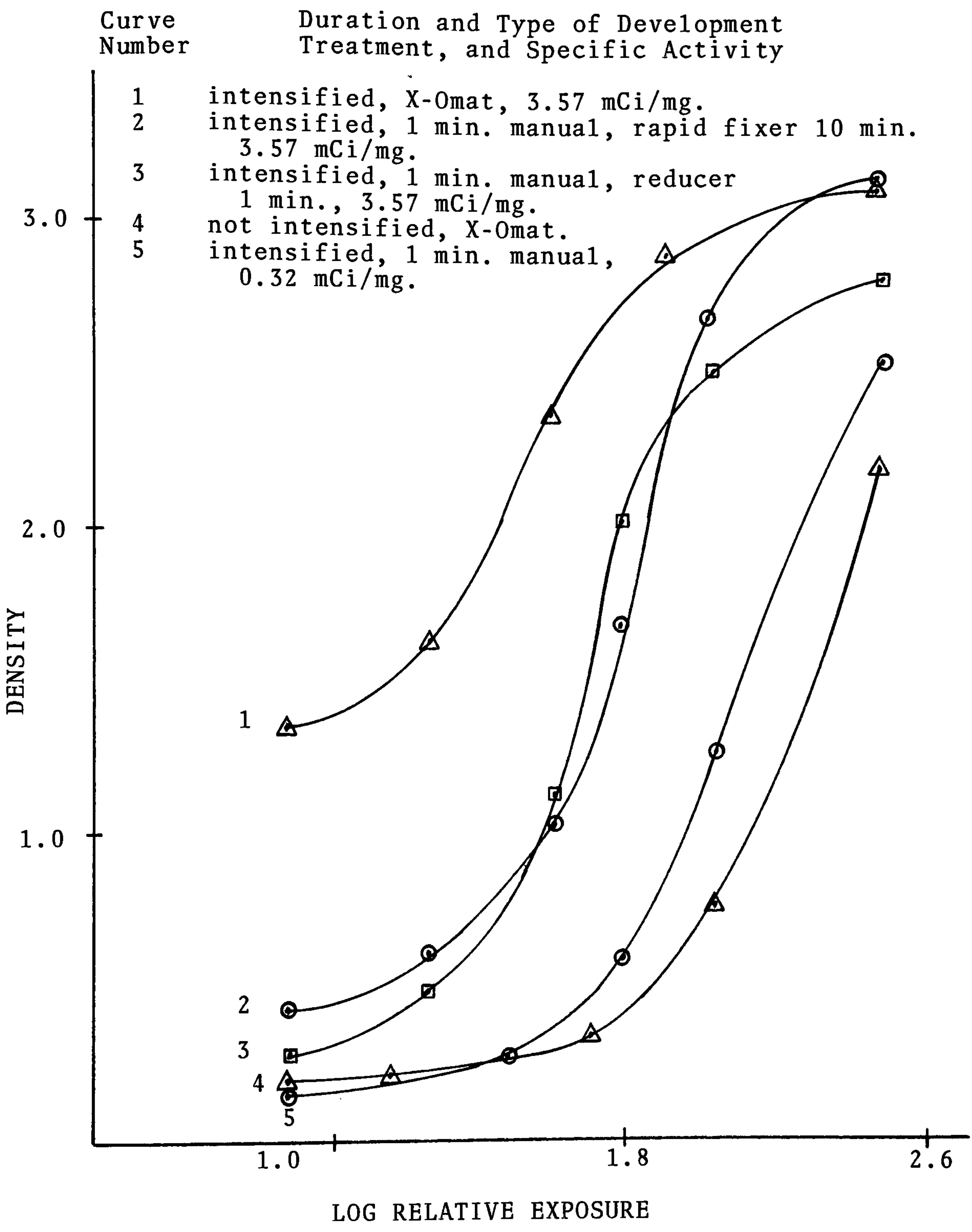


Figure 14. Characteristic Curves of Intensified 30% Exposed Images Processed in X-Omat or Manually for 1 Min. Development Time, Treated in Reducer for 1 Min. or Rapid Fixer for 10 Min., Radioactivated in 3.57 or 0.32 mCi/mg. Thiourea, and Exposed for Two Hours

The optimum development time for autoradiographic intensification was selected to be one minute based on the photographic parameters given in Table 11. Developing for one minute resulted in a decrease in the fog density from 1.31 to 0.58 and an increase in the contrast from 1.7 to 3.0. All other parameters did not change significantly.

Since the use of LSA thiourea (0.32 mCi/mg.), as discussed in section III, and decreased development of the original film, as discussed above, each separately produced a decrease in the intensified fog density, the combination of the two should have caused an even larger decrease. This, in fact, did ocur as indicated by a decrease from 0.58, for the intensified one minute developed image activated with HSA thiourea, to 0.12 for the same image activated with LSA thiourea. However, the large decrease in all other parameters was not acceptable. This low level of intensification is probably due to less radioactive silver sulfide being formed because of less developed silver and more available nonradioactive thiourea.

The combined effects of decreased development (1 min.) and treatment in subtractive reducer (Figure 12, curves 2, 3, and 4) or extended fixation in Kodak rapid fixer (Figure 13, curves 2, 3, and 4) also produced a decrease in the intensified fog density. This was possibly due to less emulsion and development fog being produced in the original, in the first place, and a decrease in whatever fog was produced as a result of treatment in the reducer. This decrease in the already developed fog was accomplished by pretreatment with the subtractive reducer or extended fixation with Kodak rapid fixer prior to development. A comparison of the effectiveness of the subtractive reducer (curve 3) and the extended fixation (curve 2) is shown in Figure 14. The pretreatment with the reducer produced a lower intensified fog density (0.26 versus 0.40), but a lower maximum density (2.79 versus 3.06), while contrast, speed, and resolution were unchanged. The reducer-treated one minute developed original may produce the better quality image because of the lower fog density. However, extending the fixation period beyond 10 minutes may also possibly be used to obtain a lower fog density.

The characteristic curves for the intensified images of the one minute developed images obtained by activating in LSA (curve 5) and the X-Omat processed image obtained by radioactivating in HSA (curve 1) thiourea are also shown in Fiure 14. A visual comparison of these curves with the reducer-treated, one minute developed image activated in HSA thiourea (curve 3) and the one minute developed, 10 minute fixed image activated in HSA thiourea (curve 2), show that these latter two curves are superior. A comparison of these two curves with a fully developed (X-Omat), 30% exposed image pretreated in subtractive reducer should have followed. However, the lot of thiourea used for these development experiments had been consumed before this could be completed. Since considerable variation from one lot of thiourea to another was experienced in earlier experiments, this intensification with a different lot of thiourea was not completed. If done, a valid comparison could not have been made.

VIII SUMMARY

The fog level of an autoradiographically intensified image was decreased by the following methods:

1. producing a more proportional conversion of silver to silver sulfide during the radioactivation by decreasing the specific activity of the thiourea solution.
2. decreasing the amount of fog silver in the developed original image by treating it in the Kodak rapid fixer prior to radioactivation, and
3. minimizing the amount of fog formed in the original image by developing for a shorter period of time than recommended for full development.

These methods of decreasing autoradiographic fog also resulted in a slight loss in the ASA speed, in most cases. Manually developing the underexposed image for one minute, decreased the fog density by 50% without causing any decrease in the ASA speed. Decreasing the specific activity of the 0.5 mCi/mg. thiourea solution from 3.57 mCi/mg.

to 0.32 mCi/mg. was more effective in decreasing the level of intensified fog, but at the expense of a loss in the ASA speed. The results further idicated that Kodak rapid fixer is an effective photographic reducer and can be used as an alternative to a subtractive reducer - consisting of potassium ferricyanide and sodium thiosulfate - to decrease the intensified fog associated with AIM.

The use of potassium ferricyanide and hydrogen peroxide as oxidizing agents for the silver did not produce a more proportional intensification. The addition of potassium ferricyanide to the thiourea solution resulted in a more disproportional intensification, while hydrogen peroxide produced little or no change in the intensification.

The reaction of gelatin with sulfur-35 thiourea, as a source of autoradiographic fog, was ruled out. It was concluded that the fog silver in the intensified image was due to radioactivation of the non-image silver, which can be minimized by either preventing its formation or removing it if allowed to form.

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APPENDIX

Calibration of the Aluminum 11 Step Wedge

The 11 step wedge consisted of high purity aluminum, with each one-half inch wide step increasing by three millimeters. It was calibrated according to the method used by Gorski and Plewes[1].

Kodak film type XG-1 was exposed on both sides through a calibrated step tablet in a cassette containing an intensifying screen. The step tablet contained 21 steps with a density range from 0.05 to 3.05 in 0.15 increments. The cassette was designed such that it protected the film from X-rays with lead glass but permitted the film to be exposed to the light emitted from the intensifying screen. This light was attenuated by the 21 step tablet prior to its incidence on the film. Shown in Figure 15 is the Density versus Log Relative Exposure curve for this exposure. The densities are listed in Table 15.

The aluminum 11 step wedge was imaged at 80 KVp onto Kodak film type XG-1 in a cassette containing the same intensifying screen. The corresponding densities are listed in Table 16 and the Density versus Step Number curve is shown in Figure 15.

Using the calibrated 21 step tablet as a reference, the difference in the log exposure corresponding to each two consecutive steps of the aluminum step wedge was determined. As an example, the difference in log exposure between step 1 and 2 of the step wedge will be determined. The densities corresponding to step 1 and 2 of the aluminum wedge were found on the step wedge curve. These same densities were located on the step tablet curve and the corresponding difference in the log exposure was determined. This log exposure difference was then used as the difference in the log relative exposure between steps 1 and 2 of the aluminum step wedge. The dotted lines in Figure 15 explain this method. The log exposure difference for each two consecutive steps was determined in this manner. The log exposure value for step 0 was arbitrarily set at 3.00 and all other log exposures were determined relative to this value. The log relative exposure values are also listed in Table 16.

APPENDIX REFERENCE

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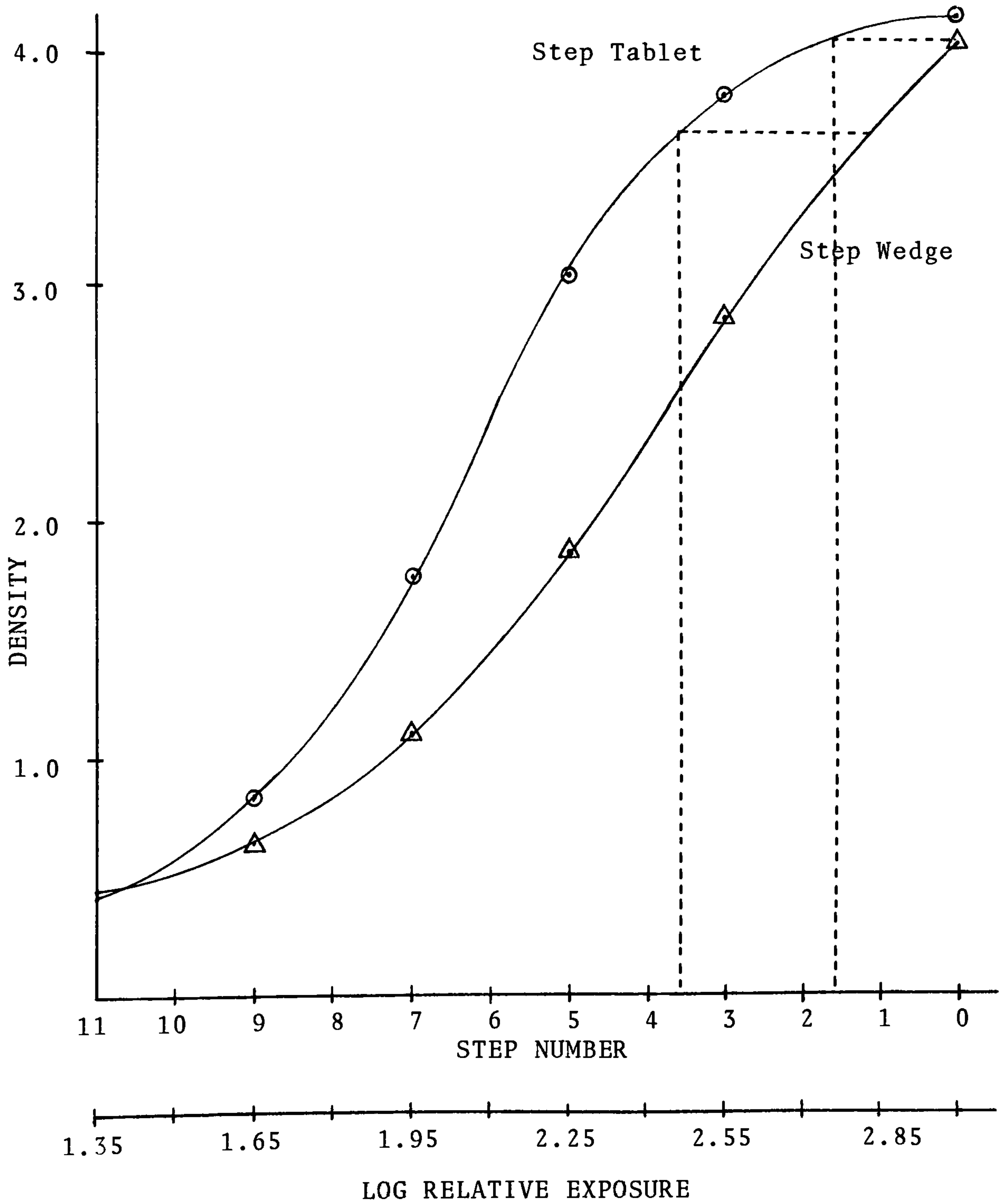

Figure 15. Density Versus Step Number/Log Relative Exposure for the Exposed Step Tablet and Step Wedge

TABLE 15

Density and Log Relative Exposure Values
for the Sensitometric Exposure

Step Number*	Density	Log Relative Exposure
0	4.16	3.00
1	4.13	2.85
2	4.03	2.70
3	3.83	2.55
4	3.54	2.40
5	3.04	2.25
6	2.46	2.10
7	1.77	1.95
8	1.20	1.80
9	0.84	1.65
10	0.56	1.50
11	0.40	1.35

* Only data for steps 0 thru 11 are shown for the 21 step tablet

TABLE 16

Density and Log Relative Exposure Values
for the Aluminum Step Wedge

Step Number	Density	Log Exposure Difference	Log Relative Exposure
0	4.06		3.00
1	3.65	0.30	2.70
2	3.30	0.15	2.55
3	2.86	0.12	2.43
4	2.33	0.12	2.31
5	1.86	0.09	2.22
6	1.43	0.10	2.12
7	1.11	0.11	2.01
8	0.85	0.12	1.89
9	0.65	0.10	1.79
10	0.52	0.09	1.70
11	0.44	0.07	1.63